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and the

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EASTERN AND CENTRAL MONTANA AS A POSSIBLE SOURCE AREA OF URANIUM [1]

FRANK C. ARMSTRONG

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ABSTRACT

Geologic settings in central and eastern Montana and in a few places in southwestern Montana are similar to the settings in areas favorable for the occurrence of uranium deposits. Several areas in Montana seem especially favorable for the occurrence of uranium.

The alkalic igneous rock province of central Montana is similar to the one of the Colorado Plateau. On the basis of this analogy, certain areas in Montana, where the lithology and (or) structure are considered favorable for the occurrence of uranium, are suggested for prospecting. Seleniferous plants, similar to those that are uranium indicators on the Colorado Plateau, might be used as an aid in prospecting for uranium provided that the seleniferous shale areas are taken into account.

Uranium deposits, similar to those in North and South Dakota, may be found in Cretaceous and Tertiary sandstones, lignites, and low-grade

[1] Publication authorized by the Director, U. S. Geological Survey.

coals of eastern Montana; also, the magnetite-bearing beds of certain Cretaceous sandstones may contain fossil placer deposits of radioactive black minerals. The occurrence of uranium in basal conglomerates, quartzites, and sandstones in Africa and Ontario suggests that similar basal sedimentary rocks in central Montana may contain uranium.

Many of the radioactivity anomalies in Montana found from the air are closely associated with the Eagle sandstone; others are associated with Precambrian basement rocks, Upper Cretaceous and Lower Tertiary igneous rocks, or sedimentary rocks of Jurassic and Cretaceous age. Glacial cover and lack of outcrops locally may hinder prospecting for uranium.

INTRODUCTION

THE intensive interest in uranium and the vast amount of work that has been directed during the past 10 years to the discovery of new deposits has done much to further our understanding of the occurrence of this element. One outstanding result of this work has been the realization that concentrations of uranium, far from being extremely rare, are fairly widespread and occur in many geologic environments. The idea that an empirical analysis of the distribution of known deposits and the geologic environment of each may afford some basis for directing the search for new deposits in new areas has undoubtedly been used by many prospectors. The purpose of this paper is to apply such an empirical analysis coupled with certain general theories of the origin of uranium deposits to certain geologic environments in eastern and central Montana that might contain uranium. It is hoped that this discussion will serve to stimulate interest and exploration and serve as a basis for evaluating the potential of different parts of this large area. The prospecting guides given are broad and are meant to suggest potential uranium provinces rather than specific localities.

Like all speculations about where to prospect and why, the reasons for so doing are subject to many qualifications and reservations. The "where" and much of the "why" are suggested empirically: 1) uranium has been found in eastern and central Montana; and 2) uranium has been found elsewhere in geologic environments similar to those in eastern and central Montana. The rest of the "why" is arrived at deductively by applying possible modes of origin for some uranium deposits. It is realized that the origin of many uranium deposits is an extremely controversial subject, and that any conclusions deduced from a postulated mode of origin are only as good as the original premises.

The ideas in this article are a product of two years' reconnaissance for uranium in Idaho, Montana, Oregon, and Washington and were first written up in a preliminary form in the spring of 1954. Since that time extensive prospecting for uranium in Montana has resulted in the discovery of a few promising deposits. The writer wishes to acknowledge the help given him by his colleagues and the benefit of discussions with them: in particular, C. E. Erdmann for information about the stratigraphy of central Montana; F. W. Cater for supplying many of the details about the Colorado Plateau uranium deposits; and S. N. Kesten for general discussions about the possibility of uranium deposits in eastern and central Montana. This work was done by

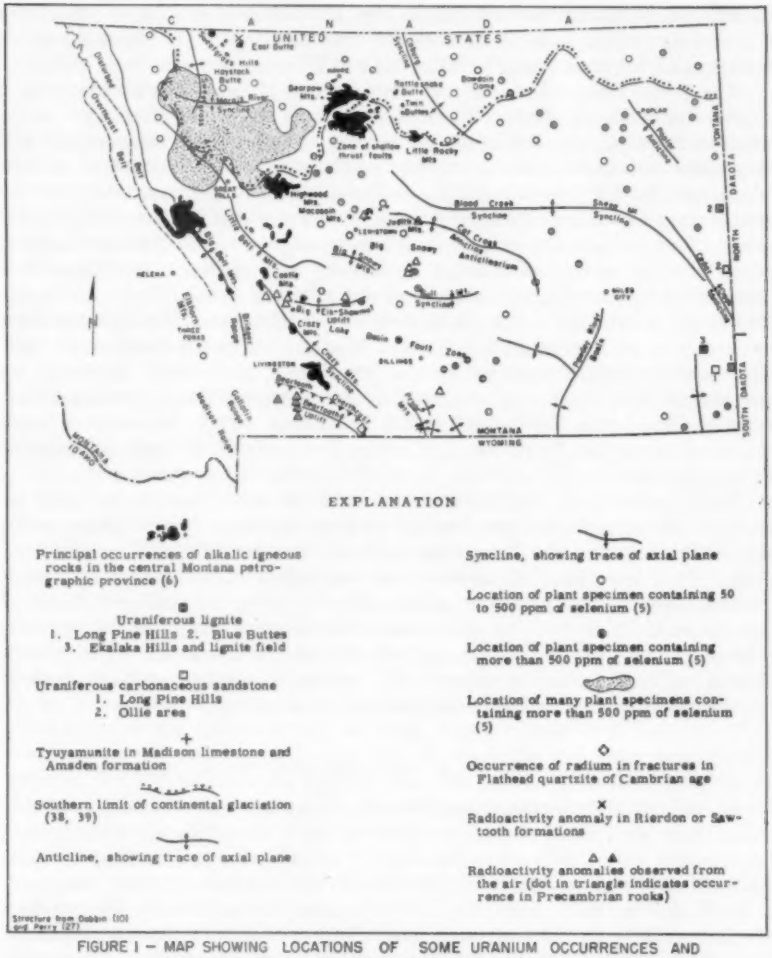

FIGURE 1 — MAP SHOWING LOCATIONS OF SOME URANIUM OCCURRENCES AND RADIOACTIVITY ANOMALIES IN EASTERN AND CENTRAL MONTANA

the U. S. Geological Survey on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission.

GENERAL GEOLOGY

The Great Plains of eastern Montana are underlain by a thick blanket of continental and marine sedimentary rocks that in places have been folded into

broad, open anticlines and synclines (Fig. 1). Most of the rocks exposed at the surface are of Late Cretaceous or Paleocene age. In places erosional remnants of rocks as young as Miocene and Pliocene are preserved (29).

The Great Plains of central and north-central Montana are underlain by a similar but thinner blanket of continental and marine sedimentary rocks. Most of the rocks exposed at the surface are also of Late Cretaceous age and in general consist of formations slightly older than those exposed in eastern Montana. In the central and west-central part of Montana many of the sedimentary formations change from marine on the east to nonmarine on the west. This change of environment of deposition gives rise to the intertonguing of marine shales and nonmarine sandstones and shales, a condition well exemplified by the Colorado shale and the Montana group (31). Deformation of the sedimentary rocks in central and north-central Montana has been somewhat more intense than in eastern Montana. Locally these rocks have been bent into sharp folds, as in the Big Snowy Mountains, or bowed up sharply, as along the Rocky Mountain front and around domal igneous mountains like the Little Rocky and Judith Mountains (11). In areas of more intense deformation, Paleozoic, and locally Precambrian, as well as Mesozoic rocks crop out.

The igneous rocks that form the cores of the domal mountains make up part of the petrographic province of central Montana (19). These rocks are of Tertiary age and have been emplaced as laccoliths, stocks, sills, and dikes. In a few places, flows have been extruded. They consist of normal granitic types as well as a suite of alkalic rocks (Fig. 1) similar to those of the Colorado Plateau. The alkalic rocks include such varieties as syenites, quartz syenites, syenite porphyries, shonkinites (melanocratic syenite with alkaline affinities), and trachyte and phonolite porphyries (fine-grained equivalents of syenite and nepheline syenite, respectively).

SOME TYPES OF URANIUM AND RADIOACTIVITY OCCURRENCES

In eastern and southeastern Montana, uranium and abnormal radioactivity have been found in many different types of sedimentary rocks that range in age from Cambrian to Miocene. "A visible but unidentified uranium mineral" occurs in sandstone of the Hell Creek formation (Late Cretaceous) in the Long Pine Hills area (Fig. 1 [2]) of eastern Carter County, southeastern Montana (38). In the same area, anomalous radioactivity has been reported from sandstone beds of the Ludlow member of the Fort Union formation (Paleocene) and from siliceous shale of the Arikaree formation (Miocene). In the Ollie area in easternmost Fallon County, uranium in near-ore concentrations has been found in thin beds of sandstone, carbonaceous sandstone, and lignitic material in the Fort Union formation. In the Ekalaka lignite field, uranium occurs in ironstone concentrations in sandstone of the upper part of the Fort Union formation (Gill, J. R., written communication, 1954).

Uraniferous lignite has been reported from the Fort Union formation in

[2] The locations of uranium occurrences, abnormal radioactivity, and radioactivity anomalies observed from the air in Montana and referred to below are shown on Fig. 1.

the Long Pine Hills, in the lower part of the same formation in the Ekalaka Hills (38) and in the upper part of the Sentinel Butte member at Blue Buttes just west of the North Dakota line (Beroni, E. P., and Bauer, H. L., written communication, 1952).

The Virgelle sandstone member (Late Cretaceous), the basal member of the Eagle sandstone, is a coarse-grained, crossbedded, massive sandstone that locally contains abundant magnetite and titaniferous magnetite, and small amounts of zircon, monazite, and garnet. This mineral suite and the littoral fauna (31) of the Virgelle suggest that the concentrations of heavy minerals probably accumulated as beach or offshore placer deposits. C. E. Erdmann (written communication, 1953) found abnormal radioactivity in the Virgelle. Because no "radioactive blacks" (uranium-bearing multiple oxides of titanium, niobium, and tantalum) have been reported from the Virgelle, it is assumed that the radioactivity is due to thorium in the monazite.

A strong "kick" on a gamma-ray oil well log has been reported from the Rierdon formation (Late Jurassic) or Sawtooth formation (Middle Jurassic) northwest of East Butte of the Sweetgrass Hills (Erdmann, C. E., written communication, 1954).

At a few places on the crest and flanks of the Pryor Mountains, tyuyamunite in ore-grade concentrations has been found recently in the Madison limestone (Mississippian) where it is overlain disconformably (30) by the Amsden formation (Mississippian and Pennsylvanian) (L. D. Jarrard, oral communication, 1956). These deposits are probably in the Mission Canyon limestone of the Madison group (4, 20). The tyuyamunite fills and coats the walls of fractures and vugs in cherty and silicified breccia zones at or near the top of the Mission Canyon. By analogy with the breccias in the Mission Canyon in the Elkhorn Mountains, the breccia probably developed as the result of solution and collapse of the Mission Canyon at the time of the development of the disconformity between the Madison and Amsden (17). Some of the tyuyamunite-bearing breccia is red. The red color suggests that some of the breccia material may be Amsden deposited in sink holes in the top of the Mission Canyon. In the same area of the Pryor Mountains, tyuyamunite also fills and coats the walls of fractures and vugs in thin beds of silicified limestone near the top of the Amsden.

Along the east front of the Beartooth uplift in south-central Montana, radioactivity anomalies have been reported by Stow (33) at several places in the Flathead quartzite (Cambrian) where it unconformably overlies Archean type rocks. At the Royse claim in this area radioactive material fills joint openings in the Flathead. Analyses by the U. S. Geological Survey show that the radioactivity is due to the presence of radium in the joint filling.

Radioactivity Anomalies Observed from the Air.—The U. S. Atomic Energy Commission has done airborne reconnaissance for radioactivity anomalies in parts of eastern and central Montana. On the anomaly maps (36) available to the writer, the Atomic Energy Commission plotted 29 positive radioactivity anomalies, and the locations of these anomalies are shown on Figure 1; in two places the anomalies are so close together that they cannot be shown individually. The five anomalies shown on Figure 1 in south-

central Montana on the Beartooth uplift are in Precambrian rocks; of this group, the four closely spaced ones represent 14 anomalies found on the west side of Boulder River. The two anomalies on the west end of the Big Elk-Shawmut uplift represent four closely spaced anomalies on the west flank of the Castle Mountains.

The westernmost anomaly on the Crazy Mountains syncline is in Upper Cretaceous and lower Tertiary volcanic rocks; and the anomaly in the Judith Mountains north of Lewiston is in Upper Cretaceous and lower Tertiary intrusive rocks. The anomaly northeast of the Pryor Mountains occurs in rocks shown as Jurassic undifferentiated on the geologic map of Montana. According to E. E. Jones (16) the anomaly is in shales of the Morrison formation (Upper Jurassic), and samples from pits dug in the shale assayed nil to 0.01 percent U_3O_8.

The rest of the radioactivity anomalies are in or near the Eagle sandstone, the basal member of the Montana group, or in the Colorado shale (both Upper Cretaceous). If the Virgelle sandstone member of the Eagle is present and contains monazite at these localities, the radioactivity anomalies are probably due to thorium in the monazite, but some anomalies may be caused by "radioactive blacks" in the Virgelle or by beds of uraniferous lignite in the Eagle.

Most of the radioactivity anomalies in the sedimentary rocks are on the margins of structurally positive areas. This structural position may reflect only the fact that monazite- or uranium-bearing Virgelle sandstone or uraniferous lignite beds are exposed at the surface on the flanks of folds.

SIMILAR URANIUM OCCURRENCES ELSEWHERE

Colorado Plateau.—The thick sequence of sedimentary rocks exposed on the Colorado Plateau is mostly of continental origin and ranges in age from Pennsylvanian to late Tertiary. Tertiary rocks are restricted largely to the margins of the Plateau. Tertiary stocks surrounded by laccoliths and other intrusive bodies (13) have intruded the sedimentary rocks and form the cores of mountains. Soda-rich syenites and andesites are exposed in the La Sal, Henry, Abajo, Carrizo, and Ute Mountains. Dikes and volcanic rocks of the Four-Corners area are subsilicic, potash-rich varieties such as nepheline syenite, shonkinite, leucite basalt, and minette (a biotite-orthoclase syenitic lamprophyre). Similar rocks also occur in the Chuska Mountains in northwestern New Mexico and northeastern Arizona. The sedimentary rocks have been tilted by monoclinal warping and around the igneous mountains they have been domed and locally turned up sharply by the intrusion of the igneous rocks. In the vicinity of the La Sal Mountains the sedimentary rocks have been faulted and folded into a series of anticlines. Considerable faulting has also taken place along the southwest edge of the Uncompahgre uplift and in the vicinity of the San Rafael swell (21).

Sandstone-type uranium ore deposits on the Colorado Plateau have been found in rocks of Permian through Late Cretaceous age. Most of the de-

posits are in continental sedimentary rocks, but a few are in marine rocks. Sandstone is the most consistently favorable host rock, but minable deposits have also been found in limestone, carbonaceous shale, shale, tuffaceous sediments, and coal. The most favorable sandstone layers are those that contain interlayered thin beds of mudstone (22). Organic matter in the sedimentary rocks was an important factor in localizing some uranium deposits.

Although at present no clear-cut structural controls are recognized for most of the ore deposits, the abundance of metals around the La Sal Mountains shows a gross zonal pattern that is independent of the stratigraphy of the host rocks (38, 37). Gold and copper occur in the La Sal Mountains, uranium-vanadium deposits to the east and south of the mountains, and a generally non-uraniferous copper-rich zone between the two. At a few uranium deposits, structural controls are recognized: the Monument No. 2 and Rajah ore deposits are related to faulting, and southeast of the La Sal Mountains some deposits are found on the flanks of anticlines (Cater, F. W., oral communication, 1954).

Two types of uranium ore occur on the plateau—black ore and yellow ore. The black ore, discovered only in recent years, generally is not exposed at the surface. It contains uraninite, pyrite, galena, sphalerite, chalcopyrite, selenides, and vanadium minerals (38). In the black ore, uranium and vanadium are present in separate minerals as low-valent ions. The yellow ore consists principally of carnotite with lesser amounts of tyuyamunite, autunite, and other secondary uranium minerals. Locally the yellow ore may contain a little pyrite or be slightly copper stained. Carnotite and tyuyamunite contain both uranium and vanadium in high-valent states, and these minerals are secondary after the black ore (22).

The uranium deposits are epigenetic. Age determinations of 55 to 75 million years, with the younger determination probably being more nearly correct, have been made for uraninites from some of the deposits, and these ages are considerably less than those of the host rocks (32).

Northeastern Wyoming.—Sandstone-type uranium deposits have been found at several places in northeastern Wyoming. In the Pumpkin Buttes, Monument Hill, and Box Creek areas uranium deposits are in reddish sandstone lenses in the Wasatch formation of Eocene age (38, 39). The uranium minerals are most abundant near the gradation of the reddish sandstone into the normal buff or gray sandstone of the Wasatch. In the Devils Tower area, ore-grade uranium deposits occur in sandstones in members of the Inyan Kara group of Early Cretaceous age. Most of the deposits are in the Fall River sandstone, but some have been found in the Fuson formation and Lakota sandstone as well. Most of the uranium deposits are in an area of more intense folding on a regional monocline (38). At the Carlile mine the ore deposit is associated with carbonaceous material in interbedded sandstones, shales, and siltstones (39). The Storm Hill deposits are less important than several others in the Northern Hills, and the fact that they are on the east flank of a domal structure does not seem important to mention here.

North and South Dakota.—In southwestern North Dakota a few miles

east of the Montana State line, yellow uranium minerals have been found in the Tongue River member of the Fort Union formation, and radioactivity anomalies have been reported from the sandstone and shale beds of the Golden Valley formation of Eocene age and the Sentinel Butte and Tongue River members of the Fort Union formation (38). Uraniferous opal and carnotite have been found in the Chadron formation of Oligocene age (39). In northwestern South Dakota about 50 miles east of the Montana line, meta-tyuyamunite and meta-autunite have been found in the Ludlow and Tongue River members, respectively, of the Fort Union formation (38). In the southern Black Hills, South Dakota, sandstone-type uranium deposits occur in the Inyan Kara group, principally in the Fall River sandstone. Many of the deposits occur on structural terraces (37).

In northwestern South Dakota, strippable deposits of lignite that contain uranium in sufficient concentration to be considered ore if it were in sandstone-type ore (the U. S. Atomic Energy Commission is not buying uraniferous lignite at present), have been found in the Tongue River and Ludlow members of the Fort Union formation. Uranium-bearing lignite and lignitic shale have been found also in southwestern North Dakota in the Fort Union (38, 39). The sources of the uranium in these deposits are considered to be the slightly radioactive tuffaceous sandstones of the Arikaree formation of Miocene age, and the White River and Chadron formations of Oligocene age (37).

Blind River, Ontario.—The similarity of the geologic environment at Blind River, Ontario, to that of the uranium deposits of the Rand, Africa, prompted thorough investigation of the slightly radioactive conglomerate at Blind River and led to the discovery of large uranium deposits.

In the Blind River area, uranium deposits occur in quartz-pebble conglomerates in Precambrian Mississagi quartzite, the basal member of the Bruce series (Early Huronian), which in this area unconformably overlies Archean rocks. Subsequent to the deposition of the Bruce series the area was uplifted. Granite of unknown age that could be younger than the Bruce series crops out in the Blind River area (1, 34).

The uranium is in fine-grained uraninite and brannerite closely associated with fine-grained gold and pyrite disseminated in the matrix of the quartz-pebble conglomerate. Radioactivity is not confined to the beds at or near the base of the Mississagi quartzite and has been found in more than one bed of quartz-pebble conglomerate. Uranium content increases with depth below the surface probably owing to the leaching of the outcrops by dilute sulfuric acid formed by the weathering of pyrite (28).

POSSIBLE URANIUM-BEARING ENVIRONMENTS

Because of their similarity to uranium-bearing areas elsewhere, certain geologic environments in Montana are thought to be potentially more favorable for uranium prospecting than other areas in the State.

1) Most of eastern Montana is underlain by the Fort Union formation in

which a few sandstone-type uranium deposits have already been found in Montana and nearby in North and South Dakota. More sandstone-type uranium deposits may be found in the Fort Union, particularly in the Tongue River member, and in the Hell Creek formation. The Wasatch formation, which crops out along the Wyoming State line about 60 miles east of the Pryor Mountains, may contain sandstone-type uranium deposits similar to those at Pumpkin Buttes, Wyoming.

Additional occurrences of uraniferous lignite may be found in the Fort Union and other lignite- and coal-bearing formations such as the Hell Creek, Judith River, and Eagle. In a series of experiments Moore (24) showed that peat, lignite, and subbituminous coal removed 98 percent or more of the uranium from an aqueous solution. Some geologists think it likely that the uranium in lignites was leached by ground water from overlying tuffs or tuffaceous beds and that the lignites extracted the uranium from circulating ground water (22, 37). If this is true, then many of the lignites and low-rank coals of eastern Montana are potentially uraniferous because the distribution of the erosinal remnants of the White River and Arikaree formations, both of which are tuffaceous, indicates that these formations once covered a large part of southeastern Montana. The distribution of the erosional remnants of the Flaxville gravel, which is tuffaceous locally (8), indicates that at one time the Flaxville also had a much wider distribution, in the northeastern part of the State. This fact suggests that uraniferous lignite may be found in the Fort Peck lignite field.

2) The geologic environment of the petrographic province of central Montana is strikingly similar to that of the Colorado Plateau. In Montana a suite of granitic and alkalic igneous rocks, similar to that on the Plateau, though more potassic, intrudes sedimentary rocks of varied lithology that range from Precambrian to Late Cretaceous in age. Many of these sedimentary rocks are continental sandstones that contain interlayered shales and mudstones and abundant organic remains.

The origin of the Colorado Plateau uranium deposits is an extremely controversial subject and is far from settled. However, if the deposits are in some way genetically related to the rather unusual suite of igneous rocks on the Plateau, as thought probable by some geologists (22), then the sedimentary rocks intruded by similar igneous rocks in Montana may contain sandstone-type uranium deposits.

The Montana formations listed below are those thought most likely to contain sandstone-type uranium deposits. They are sandstones of continental or mainly of continental origin (except the Colorado shale); most of them contain interlayered shale and mudstone beds; and many of them contain abundant organic remains. Some of these formations are the westward continental equivalents of formations that are marine in the eastern part of the State. The intertonguing of the marine and nonmarine facies produces an interlayering of sandstone and shale or mudstone, a feature that characterizes the most favorable continental sandstone layers on the Plateau (22).

Formation*	Series	Approximate thickness (feet)	Remarks
Fort Union	Paleocene	4,000+	Lignite-bearing. Contains concretions locally
Tullock member (basal) of the Fort Union	Paleocene	300	Coal-bearing
Willow Creek	Paleocene	720+	Perhaps correlative with Tullock
Hell Creek	Upper Cretaceous	560	Lignite-bearing. Swamp and fluviatile deposits. Part of Hell Creek formation formerly recognized as Lance formation. Contains volcanic matter
St. Mary River	Upper Cretaceous	990	Perhaps correlative with Hell Creek
Horsethief sandstone	Upper Cretaceous	360	Believed to correlate with Lennep sandstone. Contains magnetite-bearing beds
Lennep sandstone	Upper Cretaceous	250–400	"Shallow water or fluviatile sandstone . . . contains much volcanic material" (18)
Judith River	Upper Cretaceous	500	Lignite-bearing. Contains volcanic ash
Eagle sandstone	Upper Cretaceous	200–235	Lignite-bearing. Contains cannon-ball concretions. Virgelle sandstone member at base is magnetite-bearing
Two Medicine	Upper Cretaceous	1,950	Correlative with Judith River, Clagget (marine) and Eagle sandstone above Virgelle
Colorado shale	Upper Cretaceous	1,500–2,000	Mostly marine with interbedded continental sandstones that become thicker and more numerous to the west. Contains abundant organic remains (6). In southern Elkhorn Mountains dominantly continental (Klepper, M. R., oral communication, 1955)
Kootenai	Lower Cretaceous	450–475	Coal-bearing
Morrison	Upper Jurassic	Up to 450	Contains coaly beds at top (15)

* For correlations of Montana formations see Wilmarth (42), Perry (27, 26), Billings Geological Society Guidebook (3), and Cobban and Reeside (7).

The granites in the petrographic province of central Montana probably should be prospected, for they may be uranium-rich. In writing of the rocks of the Highwood and Bearpaw Mountains, E. S. Larsen, Jr. (38) states "the potash-rich mafic rocks are not very high in uranium, but considering

their mafic character they might be so considered. In such a series granites might have high uranium content."

3) The discovery of commercial uranium deposits in a formation as widespread in Montana as the Madison limestone suggests large areas favorable for prospecting. The first areas of Madison to prospect probably should be those that duplicate as nearly as possible the geologic environment of the deposits in the Pryor Mountains. The disconformable relations of Amsden to Mission Canyon in the Pryor Mountains are found in a broad area that extends to the west and north at least as far as the Elkhorn Mountains (17, 26). Uranium deposits may not necessarily be restricted to the top of the Mission Canyon for collapse solution breccias have been found as much as 200 feet below the top of the formation in the Elkhorn Mountains and 300 feet below in the Pryor Mountains (4, 17).

The area north of Billings and at least as far west as the Big Snowy Mountains may not be as favorable an area in which to prospect the Mission Canyon because Upper Mississippian beds progressively intervene between the Armsden and Mission Canyon. The disconformity, thereby, is moved stratigraphically upwards until it disappears and the Amsden rests conformably on the Heath shale of Upper Mississippian age (30, 20). The Amsden disconformably overlying the Mission Canyon also is considered to be a likely host rock for uranium and should not be overlooked in any prospecting program.

4) The occurrence of abnormal radioactivity in the Flathead quartzite immediately suggests the possibility of finding uranium deposits in Montana similar to the deposits at Blind River, Ontario, or the Rand, Africa. All such quartzites and conglomerates in even remotely similar geologic environments may well be potential host rocks for uranium deposits and probably warrant diligent prospecting. At several places in Montana the Neihart (Precambrian) and Flathead quartzites rest unconformably on Archean type rocks, as does the Mississagi quartzite at Blind River. Davidson (9), in mentioning the broad geologic environments of several large uranium-bearing areas, noted that in each area "the old granitic [Archean type] rocks have undergone domal uplift since the covering blanket of sediments was deposited, due presumably to [the intrusion of] 'younger' granites which are not always exposed," and he wondered "whether these similarities are more than coincidences." At several places in central Montana the Archean type rocks have been domed by the intrusion of Tertiary granitic and alkalic rocks, and around these intrusions the Neihart and Flathead have been bowed up sharply. By analogy with other localities these areas of domal uplift may be favorable places for the occurrence of uranium deposits and the basal quartzites may be the logical host rocks.

In the Little Belt Mountains (40, 41) both the Neihart and Flathead quartzites unconformably overlie Archean rocks and locally the Flathead has been intruded by the post-Cretaceous Wolf and Barker porphyries. In the Castle Mountains (40) the Flathead occurs near the Castle granite and locally has been thermally metamorphosed by it. The Castle granite is one of the granites associated with the alkalic petrographic province of central

Montana and according to Larsen (38) may be uranium-rich. Small amounts of silver, copper, and gold, common associates of uranium, have been mined from the Little Belt and Castle Mountains, and the geologic setting together with the presence of the mineral associates suggest that uranium deposits may be present.

The Flathead also lies on Archean type rocks in the Bridger Range and along the north front of the Beartooth uplift south and east of Livingston (14, 25, 23). Near the Wyoming State line south of Livingston, areas of Flathead resting on Archean type rocks are preserved in down-dropped fault blocks. In the Madison and Gallatin Ranges south of Three Forks, Peale (25) has mapped several miles of the Archean-Flathead contact, and at one place southwest of Three Forks he shows a small area of Archean type rocks unconformably overlain by arkosic sandstones and conglomerates of the Belt series. Several less extensive areas where the Flathead quartzite rests on Archean type rocks are known in southwestern Montana (29).

5) The abnormal radioactivity found in the Virgelle member and the radioactivity anomalies observed from the air over the Eagle sandstone or very close to it, point to the Virgelle as a possible source of uranium even though the abnormal radioactivity of one sample of Virgelle was thought to be due to thorium. The magnetite-bearing beds of the Horsethief sandstone likewise appear favorable for the occurrence of uranium.

CONDITIONS AFFECTING PROSPECTING FOR URANIUM

A few ideas have been presented to suggest geologic environments in eastern and central Montana that are thought favorable for the occurrence of uranium deposits. The possibility must be faced, however, that there may be very little uranium in the eastern and central parts of Montana. The writer hesitates to attribute the scarcity of known uranium occurrences to the lack of prospecting, but nevertheless such a lack may in part explain this scarcity.

The thick glacial deposits that cover a large part of northern Montana east of the Rocky Mountains (Fig. 1) (2, 12) may also help to explain why so few uranium occurrences have been reported. In many places, such as the area of the Sweetgrass arch north of Great Falls (Fig. 1), the glacial deposits almost completely blanket the other sedimentary rocks and would mask or greatly dilute the radioactivity anomaly of a uranium deposit in the underlying rocks. In such areas windows eroded through the glacial deposits may afford valuable data when adequately prospected, and drill holes and drill-hole cuttings should be examined for abnormal radioactivity.

Other factors that one might think would contribute to the lack of known uranium occurrences are leaching of surface deposits during a more humid climate in the past and the absence of characteristic Colorado Plateau canyon and mesa topography, which might have exposed favorable host rocks more extensively. However, leaching and absence of plateau-type dissection are unlikely to be large contributing factors, because a short distance south, in Wyoming, and southeast, in South Dakota, where the climate could never have been very different from that of Montana, secondary uranium minerals are common in a terrain no more dissected than eastern and central Montana.

AIDS TO PROSPECTING

Although the distribution and abundance of selenium-concentrating plants have been used successfully as a guide to ore in some places on the Colorado Plateau (5), it may be impossible or extremely difficult to use this prospecting method in Montana even though seleniferous plants are abundant and widespread (Fig. 1) (35). If this method is used, the results will have to be interpreted with great care because a large part of the plains of Montana is underlain by seleniferous marine shales, such as the Pierre, Bearpaw, and Colorado shales, which support seleniferous vegetation but are not ordinarily favorable host rocks for uranium deposits.

Radioactivity surveys of oil wells and deep seismograph shot holes may prove a useful tool in prospecting for uranium in the plains of Montana.

Uranium analyses of water samples from water wells in the plains of Montana may also help to delimit areas for intensive prospecting. Analyses of surface waters have been used for uranium prospecting, but, to the writer's knowledge, no attempt has yet been made to use well waters for this purpose.

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THE SOLUBILITY OF SOLIDS IN GASES

GEORGE W. MOREY

ABSTRACT

That gases can dissolve solids has been known since the work of Hannay in 1879–1881. The historical and theoretical aspects of this phenomenon are discussed, and the literature of the subject is summarized and reviewed. Some previously unpublished results from the Geophysical Laboratory on the solubility of UO_2, Al_2O_3, NiO, Nb_2O_5, Ta_2O_5, BeO and GeO_2 at 500° C, 1,000 bars steam pressure, are included.

HISTORICAL

THE idea of the dissolving of a solid substance by a gas may sound paradoxical, but the reality of such a solubility has been demonstrated repeatedly. Probably the first to call attention to this phenomenon was Hannay (20, 21, 22, 23), who showed that a solution of $FeCl_3$ in either alcohol or ether deposits some $FeCl_3$ just below its critical point, but that above the critical point the gas redissolves the solid. The solubility in alcohol (critical temperature 234.46° C) of KI, KBr and $CoCl_2$ was found to be uninterrupted by passage of alcohol through the critical state. At 320° C the beautiful blue color characteristic of $CoCl_2$ solutions remained unimpaired, and the absorption spectrum in alcohol solution was the same at 300° C as at 15° C. Wood (80) found that when either iodine or bromine was dissolved at 300° C in sufficient gaseous CS_2 (critical temperature 273° C) it gave the absorption spectra, not of gaseous iodine or bromine, but of solutions of these substances. Hannay also found that a metal could be transported through a gas. He placed mercury and either sodium or potassium in separate parts of a tube and introduced hydrogen at high pressure. The alkali metals were quickly transported through the hydrogen to the mercury, with formation of amalgams.

Villard (75) showed that iodine is dissolved both by oxygen and by carbon dioxide, and that solutions of iodine in carbon dioxide showed the spectrum of dissolved iodine, not of iodine vapor. Talmadge (73) studied the solubility of naphthaline in ether and of naphthaline and camphor in acetone, methyl alcohol and ethyl alcohol by a distillation method, in all cases at pressures less than 1 atmosphere, and found transport values substantially greater than corresponded to the vapor pressure of the solid, with the unexplained exception of camphor in ethyl alcohol. Ingerson and Morey (24) discussed the nature of the ore-forming fluid, and Morey (35) the solubility of solids in water vapor. Booth and Bidwell (4) gave a valuable summary of solubility measurements in the critical region, and Verhoogen (74) discussed the thermodynamics of a magmatic gas phase.

DISCUSSION OF THEORY

The elementary principle governing a mixture of two gases is Dalton's law, which states that the pressure of a gaseous mixture is equal to the sum of the pressures of the individual gases existing each by itself with the same temperature and volume as the mixture. From this one would expect that the amount of a solid substance to be contained in a coexisting gas of different composition would be that corresponding to the vapor pressure of the solid, an insignificant amount in the mixtures discussed in this paper. When the gaseous component (assumed to be insoluble in the solid substance) is at high pressure, the effect of that pressure is to increase the vapor pressure of the solid in accordance with a principle first enunciated by Gibbs (14; 15, pp. 154–157 and eq. 272), which has come to be known as the Poynting (54) effect. The proportional change in vapor pressure is equal to the ratio of the specific volume of the substance in question in the solid state to its specific volume in the gaseous state, or

$$\frac{dp'}{dp''} = \frac{\bar{v}''}{\bar{v}'}$$

As stated by Gibbs, this quantity is in general small. As calculated below, for the system air—CO_2 at − 150° C at 200 bars it amounts to 6 percent; at 600° C, 1,000 bars of steam pressure about doubles the vapor pressure of quartz.

The actual amount of solid material found in a compressed gas is always greater than that predicted from Dalton's law, often enormously greater. Webster (77) found that at − 35° C the amount of H_2O in air compressed to 200 atm. in equilibrium with ice is 3.14 times that predicted from its vapor pressure. Webster (78) also found impressive solubilities of solid CO_2 in compressed air. The amount of CO_2 in the gas at − 150° C and 200 atm. is 1,481 times that predicted by Dalton's law!

The constitution of these gaseous solutions is an interesting problem concerning which we can only speculate. Fundamentally we know that the chemical potential of the material at constant temperature must be the same in the solid and gaseous phases: in the solid the chemical potential is a function of pressure only; in the gas it is a function of pressure and composition. But obviously the usual approximations for the change in chemical potential with composition are inadequate. The basic problem is essentially the same whether the gaseous phase is a mixture of two gases (2, 32), in which, in general, the equilibrium pressure of a real gas in a mixture may be far from equal to the product of the mole fraction and the total pressure; or whether the denser phase is a liquid, which exhibits a real solubility in a gas phase, that is, the amount of the liquid species in the gas is greater than would be calculated from its vapor pressure; or whether the denser phase is a solid. Several authors (12, 55, 56, 79) have discussed the problem by the methods of statistical thermodynamics.

The many binary systems in which the solubility of a solid in a gas has

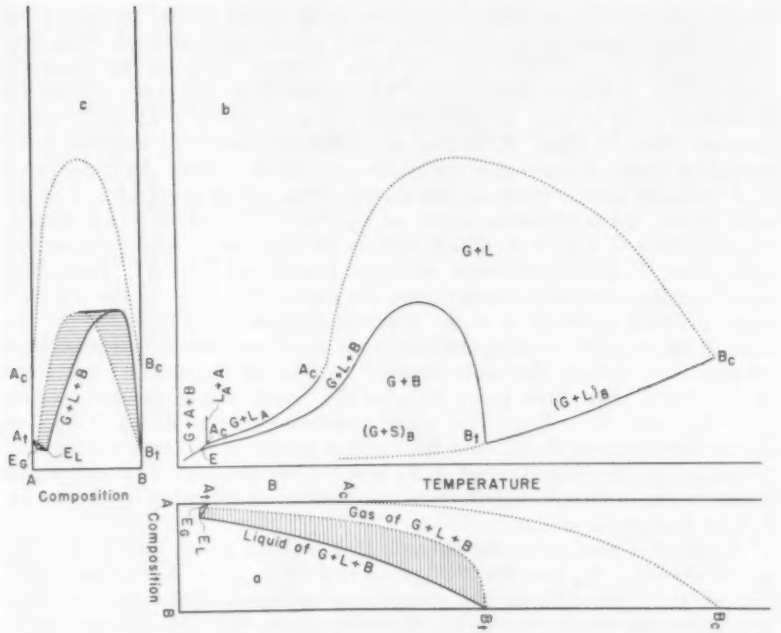

FIG. 1. Projections of a solid pressure-temperature-composition model for the case that no compounds are formed, there is no liquid immiscibility, and the solubility curve does not intersect the critical curve. Figure 1a is the projection of the solid P-T-X model on the T-X plane. A_t is the triple point of A, the curve A_tE_L the solubility curve of solid A, ending at the binary eutectic, and E_G is the gas phase in equilibrium with the liquid E_L. The curve E_LB_t is the ordinary solubility curve, which ends at the triple point of B. Figure 1b is the projection of the solid P-T-X model on the P-T plane. The curve $L_A + A$ gives the change of the melting point of A with pressure; the curve $G + L_A$ the vapor pressure curve of pure A, ending at its critical point A_c; and the intersection of these two curves is the triple point of A. The curve $G + A + B$ goes from the eutectic E to lower pressures and temperatures; the curve from the triple point of A to the eutectic E is $G + L + A$. From the eutectic the three-phase curve $G + L + B$ rises to a maximum, then falls to the triple point of B. From B_t to lower temperatures, indicated by a broken line, is the vapor pressure curve of B; from B_t to higher temperatures is the vapor pressure curve of liquid B, $(G + L)_B$, which ends at the critical point of B, B_c. The curve A_cB_c is the critical curve joining the two critical points.

been demonstrated may be divided into two classes,[1] namely: (1) those in which the solubility curve does not intersect the critical curve and (2) those in which the solubility curve intersects the critical curve. The first of these is represented in Figure 1, which is a projection of the three-dimen-

[1] Complications caused by the formation of compounds or by the formation of immiscible liquids are omitted from consideration.

sional pressure-temperature-composition solid model of the system A—B on the temperature-composition (Fig. 1a), pressure-temperature (Fig. 1b), and pressure-composition (Fig. 1c) faces. Figure 1b gives the pressure-temperature or P-T relationships. The curve $(L + S)_A$ gives the change in melting point of solid A with pressure, the curve $(G + L)$ is the vapor pressure curve of liquid A, ending at the critical point A_c, and the intersection of these two curves is the triple point of A. From the triple point of A to lower temperatures go the curves along which crystalline A is the solid phase: the sublimation curve of ice, $(G + S)_A$, which is not shown, and the curve $G + L + A$, which ends at the eutectic E, where the second solid phase, B, is in equilibrium with gas, liquid, and solid A. From E to lower temperature and pressure goes the curve $G + A + B$, the common vapor pressure curve of $A + B$. The three-phase curve $G + L + B$, the vapor pressure curve of saturated solutions of B rises from E to a maximum pressure, then falls to the vapor pressure of B at the melting or triple point of B. From this triple point go also the unary vapor pressure curves $(G + L)_B$ and $(G + S)_B$. The vapor pressure curve of liquid B ends at the critical point of B, B_c, and the critical points of the two components are joined by the critical curve A_cB_c, which gives the critical pressures and temperatures of mixtures of the two components as indicated by the curves A_cB_c in Figures 1a and 1c.

In the P-T projection the eutectic E is a single point, but in the T-X projection (Fig. 1a) and the P-X projection (Fig. 1c) it is represented by two points. One of these, E_G, gives the composition of the gaseous phase in equilibrium with the liquid, E_L. To make the relationships more evident, the content of dissolved solid in the gas is represented as greater than in any water solution known to me. A_tE_G and A_tE_L thus are the gas and liquid compositions which coexist with solid A down to the eutectic. The curves E_GB_t and E_LB_t are the gas and liquid curves of the three-phase equilibrium $G + L + B$, which go from the eutectic to the triple point of B.

The region in which solubility of a solid by a gas is exhibited is the region $G + B$ in Figure 1b. If the gaseous pressure at constant temperature is increased from the region $G + B$ to that of the three-phase curve $G + L + B$, the solid melts, and at higher pressures solid disappears and the mixture is in the region $G + L$. If the mixture is in the region above the curve $G + B$, the much greater solubilities of B in gaseous A will be from a liquid, an unsaturated solution of B in A.

This type of system is found in many cases of soluble salts in water. Typical is H_2O—NaCl, shown in Figure 6. The maximum pressure on the three-phase curve is 414 bars at 640° C. This system is discussed in detail subsequently.

The second type of system, in which the critical curve is intersected by the solubility curve, is illustrated in Figure 2. This also is a projection of a three-dimensional P-T-X model of the system A—B on the P-X, P-T, and T-X planes. Figure 2a is the temperature-concentration, (T-X), projection. It shows the solubility of B in liquid A, the curve $G + L + B$, decreasing in the region of the critical temperature of component A, and the solubility of component B in gaseous A increasing in the same region,

until the two curves merge with a vertical tangent. Here the two phases G and L become identical in composition and properties and there is a critical end point $(G = L) + B$.[2] In the P-T projection (Fig. 2b) is shown the curve $G + L + B$ terminating at this critical end point p, which represents the intersection of the pressure-temperature curve of the three-phase curve with the critical curve, a section of which is shown as $A_c p$.

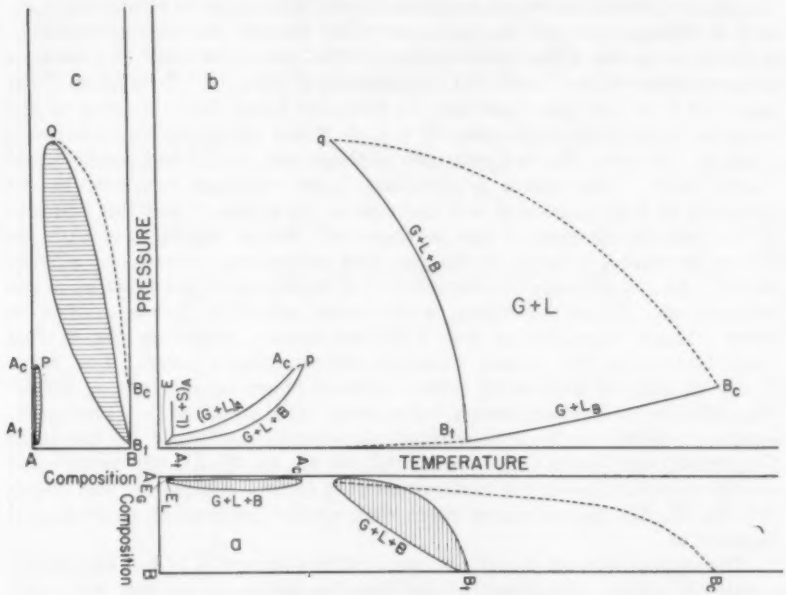

FIG. 2. Projections of a solid pressure-temperature-composition model for the case that no compounds are formed, there is no liquid immiscibility, and the solubility curve intersects the critical curve, cutting it into two parts, $A_c p$ and qB_c. Figure 2b is the projection of the solid P-T-X model on the P-T plane. From the eutectic E the three-phase curve $G + L + B$ rises until it cuts the critical curve at p; in the T-X projection the gas and liquid curves merge with a vertical tangent. From B_t along the three-phase curve $G + L + B$ pressure increases with decreasing temperature until it cuts the critical curve in q, and again in the T-X projection the gas and liquid curves merge with a vertical tangent.

The melting point of B is lowered by A, as shown by the curve $G + L + B$ extending to lower temperatures in Figures 2a and 2b. The pressure required to hold the volatile component in solution rises rapidly, as shown

[2] At a critical point two coexisting phases become identical in composition and properties, as represented by the symbols $(G = L)$ or $(L_1 = L_2)$. At a critical end point two coexisting phases become identical in composition and properties, at the same time being in equilibrium with a third phase. Types of binary critical end points are: $(G = L) + S$, $(G = L_1) + L_2$, $G + (L_1 = L_2)$, and $(L_1 = L_2) + S$.

in Figure 2b, and the coexisting gas becomes of increasing density and dissolves an increasing amount of component B, until the compositions of the gas and liquid branches of the solubility curve of B merge with a horizontal tangent, giving rise to the upper critical end point q, at which again $(G = L) + S$. In Figure 2b the pressure rises along $G + L + B$ from B_t to q, but it is also possible that the upper intersection of the critical curve by the solubility curve takes place at a temperature below that of the maximum pressure on the three-phase curve, in a region in which the pressure is falling with the temperature. The former alternative, however, probably is the usual case with systems of the type water-salt. An example is the system H_2O—$Na_2O \cdot 2SiO_2$, shown in Figure 12. In Figure 2 the region of $G + S$ is found not only at pressures lower than on either of the branches of the three-phase curve $G + L + B$, but also in the region between p and q. Between the temperatures of these two critical end points liquid cannot exist. The region is bivariant; at a constant temperature the solubility of B in gaseous A is a function of the pressure, and this pressure of the volatile component can be increased almost indefinitely, with resulting increasing density of the gas, and concomitant increasing solvent power. At any pressure the solubility of B in gaseous A is a function of the temperature. Since the region is divariant, solubility curves cannot be given. Many examples of this type are known, including the system H_2O—SiO_2, and the classic example, ether-anthraquinone. The basic theory of this, as well as all other types of phase equilibrium, is Gibbs' "Equilibrium of Heterogeneous Substances" (15, p. 55–353). The application of Gibbs' methods to critical phenomena was made by his great disciple van der Waals (76). Smits (63, 64, 65, 66, 67, 68, 69) contributed greatly to our knowledge of systems showing critical end points, and Niggli (49, 50, 51, 52) has discussed them with special application to geological phenomena.

The region between p and q is one in which there can be but one phase, a gaseous phase. Problems of definition arise in connection with this phase on which there is no unanimity of opinion. The definitions preferred by me are:

1) A liquid is a non-crystalline phase which usually takes the shape of the container but does not expand to fill all the space available.

2) A gas is a non-crystalline phase which expands to fill all the space available.

3) A vapor is a special case of a gas. Definitions of it usually refer to a pure substance; a vapor is a gas at a temperature at which it can be compressed to a liquid. For a pure substance this means below the critical point; for a binary or more complex system it becomes either indeterminate or arbitrary. For example, in the case of the system H_2O—NaCl, discussed below, a continuous liquid aqueous solution extends from the eutectic to the melting point of the salt, and at every temperature a gas consisting essentially of water is in equilibrium with this liquid.

4) The term "fluid phase" is sometimes used to denote the gas phase between the critical end points p and q. It sometimes is used to indicate

both gas and liquid without distinction, sometimes to indicate a gas containing dissolved solid. It has led to confusion and is best abandoned.

EXPERIMENTAL RESULTS

Organic Systems

The systems showing solubility of solids in gas which have been studied in greatest detail are organic systems. In these the ratio of critical temperatures of the two components is not as large as in inorganic systems such as H_2O—SiO_2 or H_2O—NaCl, and neither the temperatures nor

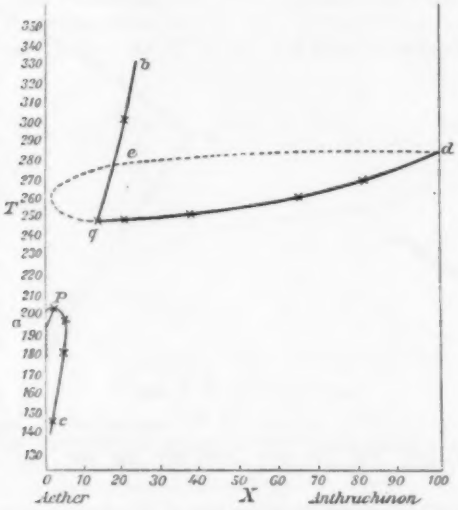

FIG. 3. Temperature-concentration projection of the system ether-anthraquinone (cf. Fig. 4). The concentration is given in mole percent of anthraquinone. The curves *cp* and *qd* are two parts of the solubility curve of anthraquinone in ether, which cuts the critical curve at *p* and *q;* the critical curve goes from *p* to the critical point of ether, from *q* along the curve *qb* to the critical point of anthraquinone. No liquid is stable between *p* and *q*. After Smits.

pressures are so large. Only a few examples of this type will be included here, including the classic example of a system showing critical end points, ether-anthraquinone. This was studied by Smits (62, 63, 64, 69) and was the first system showing critical end points of the type $(G = L) + S$ to be worked out. The melting-point or T-X diagram is shown in Figure 3. The lower part of the solubility curve ends at the critical end point *p*, at 203° C, 43 atm. Here gas and liquid become identical, each containing 4 percent by weight of anthraquinone, and the curves giving the compositions of gas and liquid coincide with a horizontal tangent. When a

tube containing 6 percent of anthraquinone was heated the point p was not reached, but liquid disappeared from the equilibrium $G + L + S$, leaving $G + S$. On further heating more and more anthraquinone went into the gas, until it all disappeared. The pressure-temperature curve is Figure 4, which shows a wide interval between the two critical end points p and q. In this interval no liquid is possible. If a mixture is compressed at a temperature either below the first critical end point, p, or above the second critical end point, q, liquid will form. But in the region between p and q

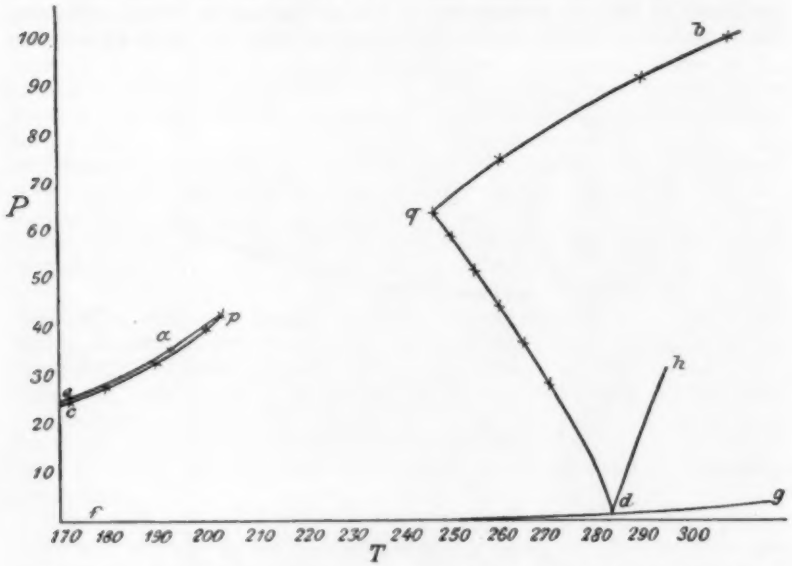

FIG. 4. Temperature-pressure projection of the system ether-anthraquinone (cf. Fig. 3). Curve ea is the vapor pressure curve of ether, a is the critical point of ether, ap is part of the binary critical curve ether-anthraquinone, which is intersected at p by the vapor pressure curve, cp, of solutions of anthraquinone in ether. Curve dq is the vapor pressure curve of solutions of anthraquinone and ether, which go from the triple point of anthraquinone, d, to the intersection with the critical curve of q, and qb is a part of the critical curve ether-anthraquinone. After Smits.

no liquid can exist, and as the gas is compressed more and more solid anthraquinone will be dissolved by the gas. The melting point of anthraquinone is at 280° C, and the second critical end point is at 247° C. At 240° C, just below the second critical end point, at a pressure of 100 atm. the gas contains 33 wt.-percent of anthraquinone. The partial pressure [3] of

[3] The partial pressure referred to here and subsequently is obtained by multiplying the total pressure by the weight fraction of the dissolved solid, in this case anthraquinone. More exact formulation would require knowledge of the mole fraction of all the molecular species present in the gas, information which we do not have.

anthraquinone is 33 atm.; the vapor pressure (25) of anthraquinone at 240° C is 0.015 atm.; so the amount dissolved in the gas is 2,200 times that to be expected from its vapor pressure.

Many other organic systems have been studied, notably by Scheffer and his coworkers (9, 10, 11, 19, 57, 58, 59, 60). One of these is ethylene (t_c = 9.9° C, p_c = 50.5 atm.)-naphthalene (m.p. 80° C), which has a first critical end point at 11° C, a second at 52° C, 174 atm. Figure 5 shows the change in solubility with pressure of naphthalene in the gas at several temperatures. "Furthermore, it is evident from these results that by raising the pressure as well as the temperature the solubility of naphthalene in supercritical ethylene increases enormously, and that, for instance, at 50° C and 270 atm. it amounts to 50 wt.-percent (18 mole %). Actually these are very concentrated solutions of slightly volatile solids in a supercritical fluid phase" (11). At 12° C and 100 atm. pressure the naphthalene

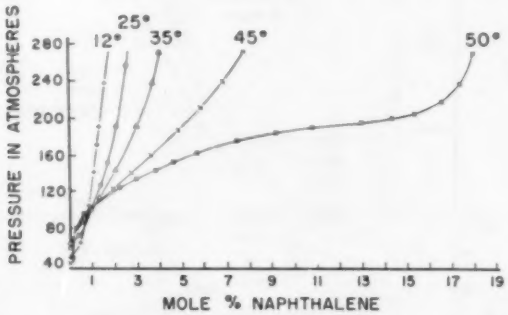

FIG. 5. Pressure-composition (P-X) sections of the system ethylene-naphthalene at several temperatures given in °C. Each curve shows the solubility of solid naphthalene in gaseous ethylene as the pressure is increased at constant temperature. After Diepen and Scheffer (11).

content of the gas is 25,500 times as great as it would be if the naphthalene had its normal vapor pressure.

Inorganic Systems

a. The Solvent is Water.—The solvent most commonly used is water, and what can happen when a mixture of salt and water is heated to a high temperature in a closed container capable of withstanding the pressure developed is a question which leads to an important and little known field of physical chemistry.

a1. There are no critical end points. As previously stated, the simplest case is that in which the solubility curve is continuous, and the critical curve is not cut by the solubility curve, illustrated in Figure 1. Large pressures of gaseous water in the presence of solid salt are not possible in this case, because the region $G + S$ is limited by the three-phase curve, and at a pressure higher than the three-phase curve the solid melts.

The water-salt system about which most is known is that with sodium chloride. Figure 6 gives the phase-equilibrium diagram of the system. The temperature-solubility diagram is fundamentally that of Keevil (27), who also determined the vapor pressure curve of the saturated solutions. The pressure-temperature curve of Figure 6 is partly based on Keevil's results, partly on results by Morey and Chen (36). The short section of the critical curve is from Ölander and Liander (53).

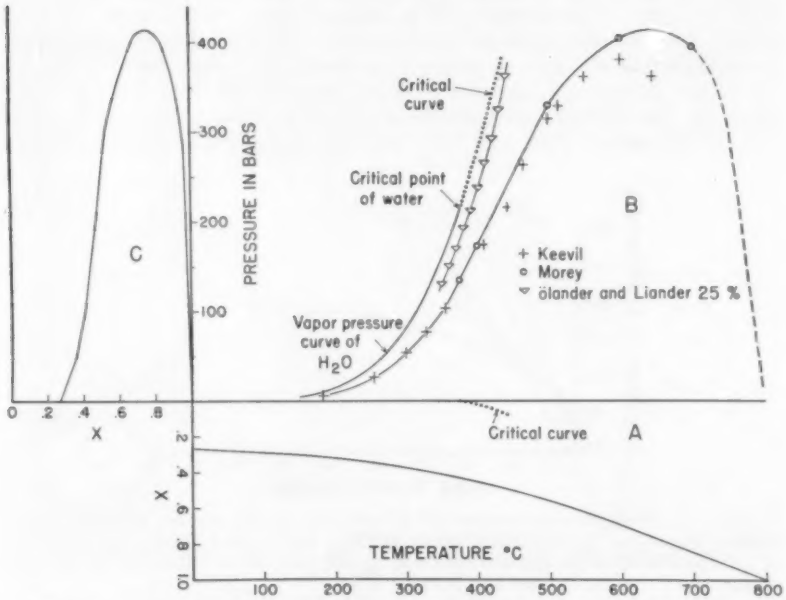

FIG. 6. The system H_2O—NaCl. In the lower diagram *A* temperature is the abscissa, weight fraction NaCl the ordinate, measured downward. The curve is based on the results of Keevil; the dotted critical curve on results of Ölander and Liander. In the diagram *B* the abscissa is temperature, the ordinate pressure, and in it are indicated the vapor pressure curve of water, ending at the critical point, the pressure-temperature curve of the three-phase equilibrium gas + solution + solid NaCl, based on the results of Keevil and of Morey and Chen. Also indicated is the vapor pressure curve of a 25% solution of NaCl in H_2O. Curve *C* gives the pressure-composition curve.

Contributions to our knowledge of the solubility of NaCl in superheated steam have been made by several investigators. Schröer (61), in his researches on the critical state found that the critical temperature of water was raised by the addition of NaCl; his results are indicated in Figure 6. Spillner (70) made measurements of the solubility of NaCl in steam at 407° C and Fuchs (13) made measurements of the NaCl content of steam in equilibrium with solutions of differing NaCl content, as well as of steam

in equilibrium with solid NaCl at 407° C. Straub (72) made similar studies. Morey and Hesselgesser (37), using a dynamic method, found the solubility of NaCl in steam at 500° C and 69 bars (1,000 psi) to be 7 parts per million; at 138 bars (2,000 psi), 14 ppm; at 276 bars (4,000 psi), 304 ppm; and at 600° C, 345 bars (5,000 psi), 539 ppm. These are superseded by the more recent results of Ölander and Liander.

Ölander and Liander (53) studied the phase-equilibrium relations of water and sodium chloride above the critical point of water, using a copper

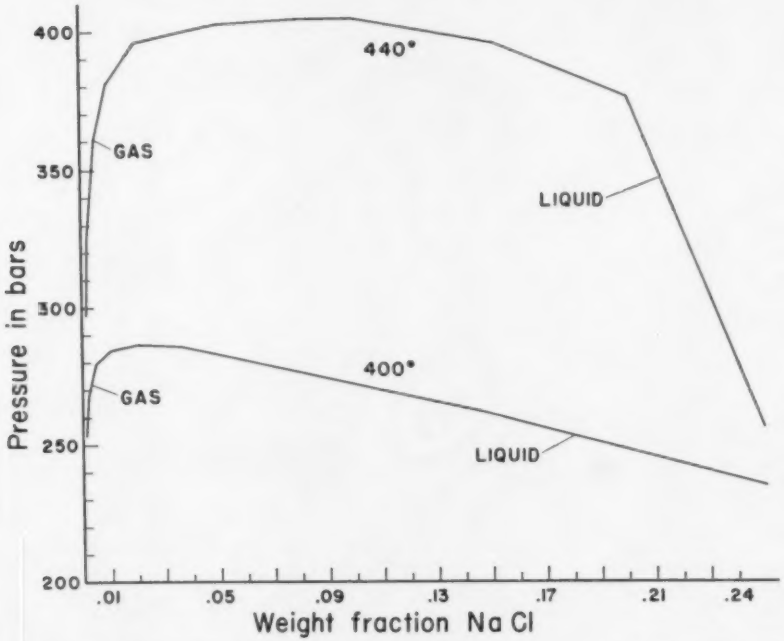

FIG. 7. The compositions of the coexisting gas and liquid at 400° C and at 440° C in the system H_2O—NaCl. At 400° C the critical point is at 286 bars, at 440° C, at 405 bars. At 400° C, 403 bars, the gas contains 5% NaCl. After Ölander and Liander.

lined steel bomb of capacity about 5 liters. Samples were let out through two capillary tubes, one ending near the top of the bomb, the other near the bottom, thus giving samples of the coexisting gas and liquid. Table 1 gives the smoothed results of the experiments, and in it is drawn the critical curve, passing through the points at which the gas and liquid become identical. Compositions below this critical curve are liquids; those above are gases. In Figure 7 are plotted the compositions of the coexisting gas and liquid phases at 400° C and at 440° C, as given in Table 1. In Figure

TABLE 1

TEMPERATURES AND PRESSURES AT WHICH A PHASE EXISTS CONTAINING THE AMOUNT OF NaCl SHOWN IN THE FIRST COLUMN.

The broken line indicates the location of the critical states, the pressures of which are given in the second line. Numbers above the broken line refer to a gas phase; those below the line, a liquid phase. After Ölander and Liander.

t =	350	360	370	380	390	400	410	420	430	440	450	460	475° C
Crit. Pr.				240	262	286	313	343	373	405	436		
0%	169	191	215										
0.005%				149	154	159	163	166	169	171			
0.01				172	178	184	190	194	198	202	205	208	
0.02				193	199	206	213	220	226	231	236	241	
0.03				206	211	218	227	234	241	248	254	259	267
0.05				220	226	234	243	252	261	269	277	285	296
0.1				234	242	253	264	275	286	297	308	318	334
0.2				240	254	268	233	297	311	325	338	352	372
0.5	167	189	213	240	260	280	301	321	341	361	379	396	
1.0	166	187	211	236	262	284	309	333	358	382			
2.0	163	184	208	234	262	286	313	340	368	396			
5.0	157	178	201	226	254	283	313	343	373	403			
10	148	169	192	216	244	284	305	338	371	405			
15	140	161	184	208	233	261	291	324	369	395			
20	133	154	176	199	222	248	277	309	341	376			
25	127	147	168	189	211	235	260	289	321	256			
sat.	113	127	141	155	169	183	198	213	227	243	258	273	297

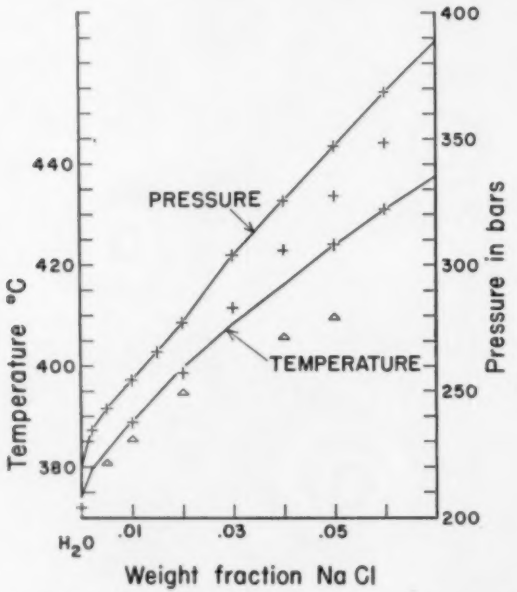

FIG. 8. The critical point of NaCl solutions. The lower curve, with scale on the left-hand side, gives the change in critical temperature with composition; the upper curve, with scale on the right-hand side, the change in critical pressure with composition. After Ölander and Liander; the points in triangles are by Schröer.

8 are plotted the change of critical temperature with composition, and of critical pressure with composition. At 400° C and 403 kg/cm² the gas contains 5 percent by weight of NaCl, so the partial pressure of NaCl is of the order of 20 atm. This is a measure of the large solubility of NaCl in superheated steam at high pressure. The vapor pressure of NaCl at 440° C, calculated from the free energy equation given by Kelley (28), is 4×10^{-8} atm., so the amount of NaCl dissolved by the gas is fifty million times that corresponding to its vapor pressure. In none of the preceding experiments was any indication found of decomposition of NaCl, with formation of HCl.

Copeland, Silverman and Benson (7) measured the density of gas and liquid phases at 385°, 390° and 396° C, with results given in Table 2. Partial molar volumes calculated from the density results were extra-

TABLE 2

COMPOSITIONS AND DENSITIES OF GASEOUS AND LIQUID SOLTUIONS OF NaCl IN H_2O

The values given are not for coexisting solutions. After Copeland, Silverman and Benson.

T (° C.)	Vapor solution		Liquid solution	
	C (% NaCl)	ρ (g/cc)	C (% NaCl)	ρ (g/cc)
385	0.12	0.22	3.00	0.50[a]
	0.15	0.24	6.74	0.60[a]
	0.19	0.26	11.96	0.70[a]
	0.44	0.32		
390	0.21	0.22	3.00	0.50[a]
	0.34	0.26	11.96	0.69[a]
	0.62	0.34		
396	0.21	0.20	3.5	0.49
	0.23	0.20	7.6	0.61
	0.30	0.24	12.4	0.69
	0.51	0.28	6.74	0.58[a]
	0.69	0.29	11.96	0.68[a]
	0.68	0.31		

[a] Estimated from pressure-density isotherms.

ordinarily high, possibly the largest ever observed. Spillner found that the steam containing dissolved NaCl had no electrical conductivity and that the dielectric constant at the critical point was 2.9, so that this water should be regarded as a "non-aqueous solvent."

The system H_2O—KCl is similar to H_2O—NaCl, with a somewhat greater solubility and lower vapor pressure. Benedict (3) described an apparatus for determining the vapor pressure of saturated salt solutions, and with it measured the vapor pressure of solutions of KCl from 250° C to 600° C. Keevil (27), using the apparatus devised by Benedict, determined the vapor pressure curve of saturated solutions, and the measurements of Morey and Chen (36) were in fair agreement, although a little higher than those found by Keevil. Neither of these made any observa-

tions on the solubility of KCl in steam. Spillner (70) made measurements of the solubility of KCl in steam at 407° C and pressures from 100 to 320 kg/cm² and at 320 kg/cm² found 0.087 weight fraction KCl.

Jasmund (26) used a method similar to that used by Gillingham (16), at 400, 450, 475 and 500° C, from 130 to 300 atm., and also a tracer technique using K^{42} from 20 to 250 atm. His results, plotted on a logarithmic scale, are shown in Figure 9. At 400° C and 300 atm. the solubility of KCl in

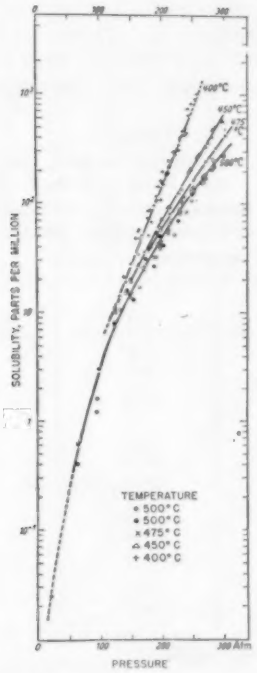

FIG. 9. The solubility of KCl in superheated steam. After Jasmund. The abscissa are pressures expressed in atmospheres, the ordinate the solubility in parts per million, on a logarithmic scale.

steam was found to be 0.017 weight fraction, at 500° C and 300 atm., 0.0278 weight fraction.

a2. There are critical end points (G = L) + S. This type of system was discussed under Organic Systems, and illustrated by the system ether-anthraquinone. To it belong the large number of difficultly soluble inorganic salts such as Na_2SO_4 and Na_2CO_3, as well as numerous oxides, such as NiO and SiO_2, and silicates.

The system H_2O—SiO_2. Probably the literature of the system H_2O—SiO_2 is more extensive than that of any other binary system with

water at high temperature and pressure. Much of the work on SiO_2 summarized by Morey and Ingerson (40) deals with liquid solutions or more complicated systems. The first reference to solubility of SiO_2 in superheated steam is to the work of Nieuwenburg and Blumendal (43, 44, 45, 46, 47). In a series of papers they demonstrated that amorphous silica could be transported through the medium of the gas phase to a crucible containing a basic oxide and fixed as wollastonite, willemite, etc., according to the basic oxide used. Nieuwenburg and Zon (48) made semiquantitative measurements on the solubility of quartz in supercritical steam. Morey (34) confirmed the transport of silica, but was not able to confirm the large values of transport. Gillis (17) also found transport of SiO_2 with formation of willemite. Greig, Merwin, and Shepherd (18) found transport of silica in sealed silica tubes containing obsidian or basalt, but no transport if the water had previously been removed. Ingerson and Morey (24) observed transport of about 50 grams of silica, which was deposited as quartz, during the course of some extended experiments at 3,000 psi. Gillingham (16) made numerous experiments which demonstrated the transfer of silica through the gas. Splittgerber (71) measured the amount of SiO_2 in the steam in equilibrium with boiler water containing 25, 100, and 200 mg per liter of SiO_2. He found with the water containing 100 mg per liter at 263° C (50 atm.) 0.07 parts per million, at 309° C (100 atm.) 0.16 ppm, and at 340° C (150 atm.) 0.90 ppm. Straub (72) passed superheated steam over precipitated silica at temperatures from 200° C to 342° C, at pressures from 67 to 420 psi. At 342° C (29 bars) he found a solubility of 2 ppm. Wyart and Sabatier (81), using the method of Nieuwenburg and Zon, at 480° C and 480 bars found for quartz 650 ppm vapor, for tridymite 1,020 ppm, for cristobalite 1,050 ppm, and for glass ± 1,200 ppm.

The researches of Kennedy (29) and of Morey and Hesselgesser greatly extended our knowledge of the solubility of silica in superheated steam at high pressures. The methods were totally different, and the results are in excellent agreement. Kennedy (30), whose results include measurements along the three-phase boundary gas + liquid + quartz, and in the two-phase field liquid + quartz, as well as in the two-phase field gas + quartz up to 560° and 1,000 bars, were made by a static method in which quartz oscillator plates were heated in closed bombs containing known amounts of water and connected to a pressure gage, and the amount dissolved determined either by the loss in weight of the quartz plate or by the change in frequency of the oscillators. Kennedy expressed his results in isobaric, isovolumetric and isothermal solubility curves. "It is further apparent from examination of the isovolumetric curves that in the critical boundary neighborhood, the property of a solvent which makes it a liquid as contrasted with the property of a solvent which makes it a gas has little or nothing to do with its efficacy as a solvent. The isovolumetric curves cross the critical boundary line separating the two-phase regions with no indication of a break or even change in slope of the solubility curve" (30).

Morey and Hesselgesser (37, 38, 39) used a dynamic method in which water was pumped through a pressure line, in which the pressure was held constant up to a throttle valve located after the water had passed through

a bomb containing quartz in the heated furnace, and the steam had cooled to room temperature. The condensed water was then weighed and the amount of dissolved SiO_2 determined by weighing as SiO_2 and evaporation with HF and H_2SO_4. The results of Morey and Hesselgesser are given in Table 3 and shown in Figure 10. Also given in Table 3 are the results of Kennedy, and it will be seen that the agreement is excellent. The column headed "Partial pressure SiO_2 bars" was "obtained by multiplying the total pressure in bars by the weight fraction SiO_2 and are not intended to convey any implication as to the molecular condition of SiO_2 in the steam, which will remain a matter of conjecture until some method of molecular weight determination under these somewhat extreme conditions is devised. Nothing is known about the extent of association of water vapor itself at

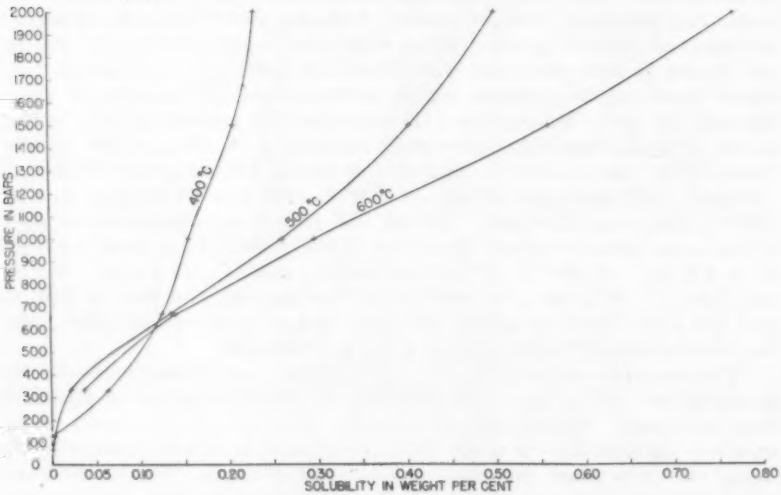

Fig. 10. The solubility of quartz in superheated steam at 400, 500 and 600° C, expressed as weight percent SiO_2 in the condensed steam. After Morey and Hesselgesser (38).

high temperature and pressure. As for the SiO_2, it is improbable that it exists as SiO_2 molecules. In all probability it exists in tetrahedral coordination as a 'silicic acid,' but speculators are incapable at present of experimental test" (38). Morey and Hesselgesser also found that in the condensed steam none of the silica appeared to be in colloidal solution.

The experimental results of Kennedy and of Morey and Hesselgesser have been discussed by several authors. Brady (5) calculated from both sets of data equilibrium constants for the formation of metasilicic acid, $SiO(OH)_2$, orthosilicic acid, $Si(OH)_4$ and the dimer $Si_2O(OH)_6$, in the gas, and concluded $Si(OH)_4$ is the volatile species at steam pressures above a few hundred atmospheres, while at lower pressures a molecule of higher molecular weight, perhaps the above dimer, exists in the gas.

Mosebach (42) made an extended discussion of the results of Kennedy and of Morey and Hesselgesser, which indicated that the silica was dissolved in the steam as $Si(OH)_4$. He derived the equation:

$$\log L = 2 \log D - 2071/T + 3.68$$

TABLE 3

THE SOLUBILITY OF QUARTZ IN SUPERHEATED STEAM AT HIGH PRESSURES

After Morey and Hesselgesser (37, 38) and Kennedy (30)

Pressure, bars	Solubility, weight percent SiO_2	Partial pressure SiO_2 bars	Kennedy
	300° C		
333–1,000	0.062		0.068–0.092
	360° C		
333	0.085		0.097
	400° C		
33	0.0001	—	—
67	0.0003	0.2×10^{-3}	—
133	0.0005	0.7×10^{-3}	—
333	0.064	0.21	0.060
667	0.126	0.84	0.142
1,000	0.155	1.55	0.173
1,500	0.206	3.1	0.217
2,000	0.231	4.6	—
	500° C		
33			
67	0.0014	0.91×10^{-3}	—
133	0.0036	3.3×10^{-3}	—
333	0.022	0.07	—
667	0.135	0.9	0.132
1,000	0.260	2.6	0.232
1,500	0.404	6.1	—
2,000	0.499	10.0	—
	600° C		
333	0.036	0.12	—
1,000	0.296	3.0	—
1,500	0.559	8.4	—
2,000	0.765	15.3	—

in which L is the solubility expressed as grams SiO_2 per kilogram H_2O, D is the density of water at the given temperature and pressure, as given by Kennedy (31), and T is the absolute temperature, which gives fair agreement with the experimental results.

Several investigators, as mentioned above, have used silica glass or

precipitated silica or other unstable forms such as tridymite or cristobalite, in their studies of the solubility of silica in steam. It is to be expected that these unstable forms would show a greater amount of dissolved silica than the stable quartz, but they cannot represent a true solubility. The criterion for testing a true solubility is that it shall represent an equilibrium condition, reversible on change of experimental condition. If an increase in pressure causes more material to pass into solution, a decrease in pressure should cause it to separate in the same thermodynamic conditions. This does not happen, and when a phase does separate it is quartz, not silica glass or tridymite or cristobalite. Morey and Hesselgesser also found much larger values with silica glass than with quartz, but in every case when the run was continued for several days the glass crystallized to quartz, and the

TABLE 4

SOME MISCELLANEOUS SOLUBILITIES IN SUPERHEATED STEAM AT 500° C, 15,000 PSI, IN PARTS PER MILLION

After Morey and Hesselgesser (37, 38) and unpublished results by Morey and Chen.

Oxides			
UO_2*	0.2	Ta_2O_5*	30
Al_2O_3*	1.8	Fe_2O_3	90
SnO_2	3.0	BeO*	120
NiO*	20.0**	SiO_2	2,600
Nb_2O_5*	28.0	GeO_2*	8,700
Sulfates			
$CaSO_4$	20	$PbSO_4$	110
$BaSO_4$	40	Na_2SO_4	4,300
Miscellaneous			
$CaCO_3$	120	sphalerite	204

* Unpublished results of Morey and Chen.
** NiO is at 30,000 psi.

observed solubility dropped to that of quartz at the given temperature and pressure. The circumstance that constant values may be obtained under a given set of experimental conditions when using one of the unstable phases does not indicate that a true solubility curve has been realized.

Miscellaneous oxide systems. Morey and Hesselgesser published the solubilities of Fe_2O_3 and SiO_2 in steam, as determined by the dynamic method, at 500° C, 1,000 bars, with results given in Table 3. Morey and Chen (unpublished results from the Geophysical Laboratory) made the measurements on UO_2, Al_2O_3, NiO, Nb_2O_5, Ta_2O_5, BeO, and GeO_2 in Table 4.

Miscellaneous salt systems. Morey and Hesselgesser measured the solubilities of several sulfates, with results given in Table 4. There was no indication in any case of decomposition of the sulfate by water with

formation of H_2SO_4. Under the same conditions, $CaCO_3$ was carried over in the steam, to the amount of 120 parts per million. An independent determination of CO_2 in the condensate gave an amount equivalent to the CaO, indicating that the $CaCO_3$ was not decomposed under these conditions.

Sulfide systems. Morey and Hesselgesser (39) made experiments by a static method at 500° C, 1,000 bars with bornite, covellite, bismuthenite, chalcocite, sphalerite, and pyrite. All except pyrite were essentially unaltered and did not show significant change in weight. For example, the bornite sample weighed 0.846 g; after the run it weighed 0.862 g and had some brownish spots and some very small recrystallized bornite on the surface. The pyrite sample weighed 1.138 g. After the run there was a

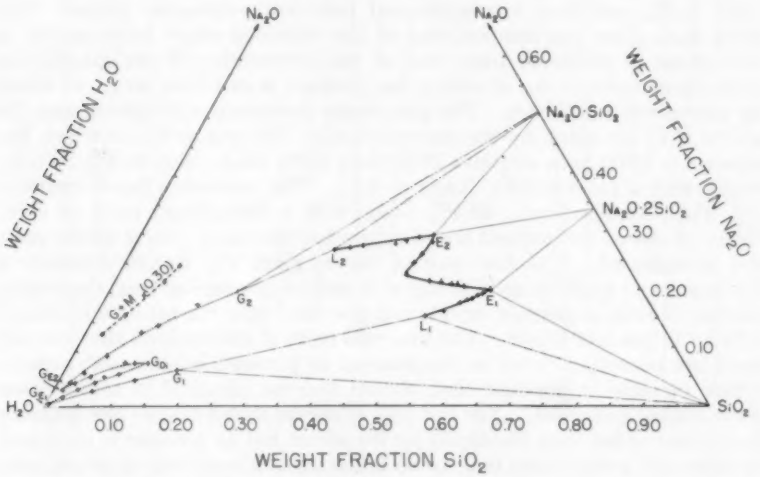

FIG. 11. The isothermal polybaric saturation curves in the system H_2O—Na_2O—SiO_2 at 400° C. The curve E_1-L_1 gives the compositions of liquid solutions in equilibrium with quartz, G_{E_1}-G_1, the coexisting gaseous solutions; and the triangle G_1-L_1-SiO_2 is the three-phase triangle at 2,500 bars pressure. G_{E_1}-G_{E_2} and E_1-E_2 are the gas and liquid saturation curves of $Na_2O \cdot 2SiO_2$. G_{E_2}-G_2 and E_2-L_2 are the saturation curves of $Na_2O \cdot SiO_2$ and L_2-G_2-$Na_2O \cdot SiO_2$ is the three-phase triangle at 2,500 bars.

strong smell of H_2S, the sample weighed 1.043 g and had a surface coating of pyrrhotite. A heavy rim of chalcocite had formed on the edge of the copper washer.

Three-component systems. In a three-component system in which compounds are formed it is only when the compounds are entirely undissociated in the gas that the ratio of the two components of lesser volatility will be the same in the gas and the liquid phases. It is probably true that in all systems, ternary and more complicated, the tendency will be for components of greater volatility to be in greater proportion in a gas than

in a coexisting liquid, and when the pressure is not great enough to form a liquid, and the equilibrium is between a gas and a solid, the amount of decomposition usually will be greater. Examples of both conditions are to be found in the system H_2O—Na_2O—SiO_2, studied by Morey and Hesselgesser (39).

Figure 11 shows the phase relations in this system at 400° C. The complete representation of the pressure-temperature-composition relations in a three-component system, in which composition is represented by X, Y, and 1-X-Y, would require a four-dimensional hyperprism with coordinates P, T, X, and Y. The relations at a constant temperature can be represented in the usual triangular prism.[4] Figure 11 is a projection of such a prism onto the triangular base: temperature is constant, so the curves L_1E_1, E_1E_2, and E_2L_2 are isothermal polybaric saturation curves. The curve E_1L_1 gives the compositions of the solutions which have quartz as solid phase at pressures from that of the intersection of the quartz and disilicate surfaces at E_1, at which the pressure is 240 bars, to L_1, at which the pressure is 2,500 bars. The gas phases coexisting with quartz and the liquids E_1L_1 are given by the curve GE_1-G_1. The gas at G_1, at which the pressure is 2,500 bars, contains 77% H_2O, 6.2% Na_2O, and 16.8% SiO_2 by weight with a ratio of SiO_2/Na_2O of 0.73. The coexisting liquid contains 35% H_2O, 15.6% Na_2O, 49.4% SiO_2, with a SiO_2/Na_2O ratio of 0.70. Figure 12 shows the gas and liquid saturation curves of quartz as the pressure is increased. The first pair of curves gives the weight fraction of H_2O in gas and liquid in equilibrium with quartz; the second pair, the weight fraction of SiO_2 in gas and liquid; and the third pair the ratio $SiO_2/(Na_2O + SiO_2)$ in gas and liquid. The first two pairs of curves show that gas and liquid are becoming closer in composition as pressure is increased, and on further increase in pressure they should become identical in composition at the critical end point. The last pair of curves shows that at low pressure Na_2O is extracted from the liquid by the steam but as pressure is increased the two solid components tend to be taken up in almost the same amount. The difference between the SiO_2/Na_2O ratio in gas and liquid is characteristic of systems in which the components differ greatly in volatility, and the circumstance that with increasing pressure the difference in ratio between the two phases may be of great significance.

The curve E_1-E_2 in Figure 11 is the isothermal polybaric saturation curve of sodium disilicate, and the curve GE_1-G_{D1}-GE_2 gives the compositions of the coexisting gases. Here again the ratio SiO_2/Na_2O differs in the gas and liquid. At the point E_1 on the boundary at 400° C between $Na_2O \cdot 2SiO_2$ and quartz, at a pressure of 240 bars, the ratio in the gas is 0.5, in the liquid, 0.741; at the boundary between $Na_2O \cdot SiO_2$ and $Na_2O \cdot 2SiO_2$, E_2, at 380 bars, the ratio in the gas is 0.32, in the liquid, 0.60. $Na_2O \cdot 2SiO_2$ has a weight percentage ratio SiO_2/Na_2O of 0.66. The gas having that ratio is in equilibrium with a liquid having the ratio of 0.732 at 1,100 bars and 400° C. It requires a water pressure of 2,200 bars to

[4] For a photograph of such a prism see Morey (33) in which are shown the T-X and P-X prisms for the system H_2O—K_2SiO_3—SiO_2.

melt $Na_2O \cdot 2SiO_2$ at 400°C. At this pressure there is not so much decomposition of the solid: in an experiment at 2,250 bars the gas had the composition 80.6% H_2O, 8.1% Na_2O, 11.3% SiO_2, with a SiO_2/Na_2O ratio of 0.584; the liquid, 33.9% H_2O, 22.4% Na_2O, 43.7% SiO_2, ratio 0.661.

In considering saturation curves such as the liquid saturation curve of $Na_2O \cdot 2SiO_2$, E_1L_2, and the gaseous saturation curve GE_1-G_{D1}-GE_2, the

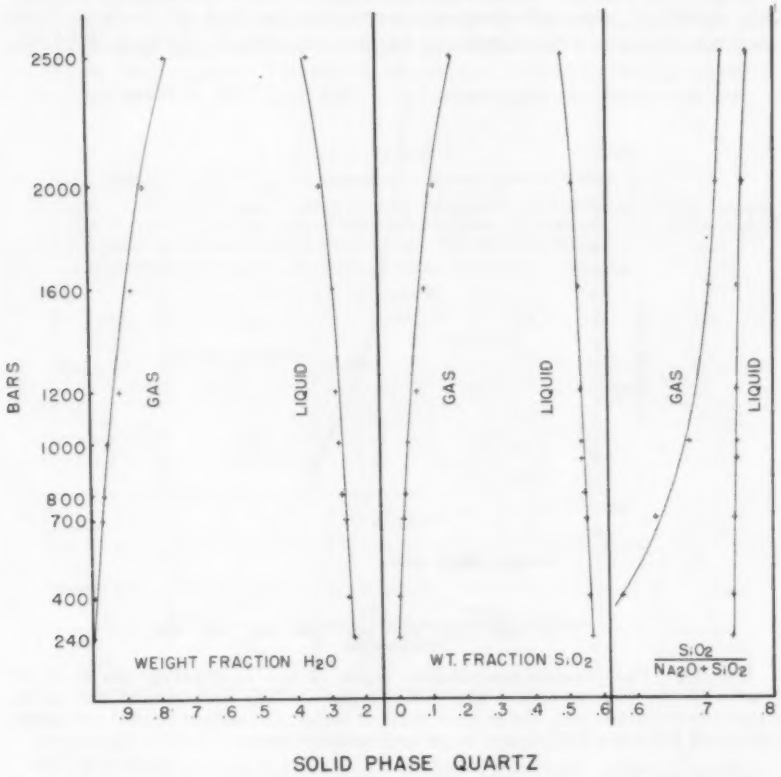

FIG. 12. The gas and liquid saturation curves of quartz at 400° C, the curves G_{E_1}-G_1 and E_1-L_1 of Fig. 11. The first pair of curves gives the weight fraction of H_2O in coexisting gas and liquid; the second pair, the weight fraction of SiO_2 in gas and liquid; and the third pair, the ratio $SiO_2/(Na_2O + SiO_2)$ in gas and liquid.

question arises as to what is the solubility of $Na_2O \cdot 2SiO_2$ in steam at a given pressure and temperature. Sodium disilicate is not incongruently soluble in either liquid or gas, because in each case the line H_2O-$Na_2O \cdot 2SiO_2$ passes through the saturation curve. But the gas having the 1:2 molecular

ratio SiO_2/Na_2O is not in equilibrium with the liquid having a 1:2 molecular ratio. On each curve the 1: 2 ratio is merely a point on the saturation curve, and the term "solubility of $Na_2O \cdot 2SiO_2$ in the gas" should not be used. This case is fundamentally different from that of Na_2SO_4, in which at the temperatures and pressures for which experimental results are available there is no indication of dissociation of the compound in the gas.

The curve E_2L_2 is the isothermal polybaric saturation curve of $Na_2O \cdot SiO_2$ at 400° C, along which the pressure changes from 380 bars at E_2 to 2,500 bars at L_2, at which the liquid has the composition 42.7% H_2O, 26.8% Na_2O, 30.5% SiO_2, ratio SiO_2/Na_2O, 0.522. The gas G_2 in equilibrium with liquid L_2 contains 61% H_2O, 19.7% Na_2O, 19.3% SiO_2, a ratio of 0.497. The line $G + M$ (.30) represents gases in equilibrium with crystalline sodium metasilicate, and one of these at 400° C, 2,000 bars contained 67.4% H_2O, 24.2% Na_2O, and 8.2% SiO_2, and had a density of 1.36. It is an extreme example of the solution of solids by gaseous water.

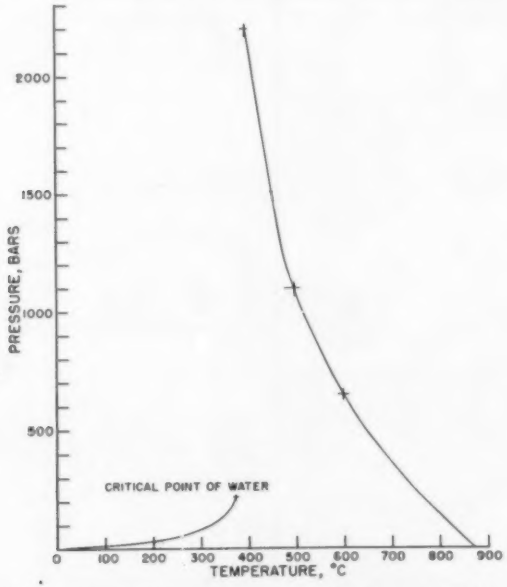

FIG. 13. The pressure-temperature curve of the equilibrium gas + liquid + solid in the binary system H_2O—$Na_2O \cdot 2SiO_2$. The first critical end point practically coincides with the critical point of water; the second critical end point is between 375° and 400° C and at an unknown pressure.

Figure 13 gives the pressure-temperature curve of the binary system H_2O—$Na_2O \cdot 2SiO_2$.[5] Sodium disilicate is very soluble in water at 200° C,

[5] The values given in this figure are from Morey and Ingerson (41), Morey and Hesselgesser (39), and unpublished results by Morey.

but the solubility curve is retrograde and $Na_2O \cdot 2SiO_2$ has practically zero solubility at the critical temperature of water; hence the first critical end point p practically coincides with the critical point of water. The melting point of $Na_2O \cdot 2SiO_2$ is lowered by H_2O, but it requires an increasingly greater pressure to hold the water in solution, and at 400° C the three-phase pressure is 2,200 bars. At 400° C, 2,500 bars, the gas contained 77% H_2O and 33% dissolved solid; a liquid of the same temperature and pressure contained 44.5% H_2O, 55.5% solid. The P-T curve is rising very rapidly with decreasing temperature, and the gas and liquid are approaching each other in composition. The second critical end point q in this binary system must be between 375° and 400° C, at some unknown high pressure. As

TABLE 5

THE DECOMPOSITION OF ALBITE AND MICROCLINE BY STEAM AT 500° C

Under "Amount dissolved" is given the total amount of material carried over in solution by the steam in parts per million, under "Mole ratio" is given the molecular composition of the dissolved material, and the last column gives the ratio SiO_2/Na_2O in the dissolved material.

Albite

Pressure, bars	Amount dissolved, ppm	Mole ratio			SiO_2/Na_2O
		Na_2O	Al_2O_3	SiO_2	
400	62	1.41	1	12.2	8.65
1,000	770	1.11	1	6.85	6.17
2,000	2,675	1.02	1	6.22	6.10

Microcline

Pressure, bars	Amount dissolved, ppm	Mole ratio			SiO_2/K_2O
		K_2O	Al_2O_3	SiO_2	
1,000	760	1.04	1	6.7	6.44
2,000	2,480	1.08	1	7.27	6.73

previously stated, the co-existing gas has in general a ratio of SiO_2/Na_2O different from that of the liquid or crystals, but if the proportion of gas is small enough this circumstance does not materially affect the coordinates of the P-T curve.

Multicomponent systems. Morey and Hesselgesser also subjected the feldspars albite and microcline to the action of superheated steam at high pressure, with results given in Table 5. The Amelia County, Va., albite used was from the same stock as that analyzed by Day and Allen (8); the microcline was from the Derry Mine, near Buckingham, Quebec. The amounts of materials dissolved were about the same for the two minerals. The measurements with albite showed a decomposition at lower pressures, which greatly decreased at higher pressures, until at 2,000 bars the albite

is carried over as such almost within the error of the experiment. At 1,000 bars, microcline showed about the same amount of decomposition as albite, but at 2,000 bars a greater amount of decomposition was indicated. It is not probable that this difference between the two feldspars can be ascribed to experimental error. It follows from the results of Table 5 that the material transported by steam at lower pressures contains alkali oxide and silica in excess of the feldspar ratio, and the material remaining contains an excess of alumina. Similar results were obtained by Morey and Chen (36) with water below its critical point.

Morey and Hesselgesser also worked with enstatite, $MgSiO_3$, and found it to be decomposed, with the extraction of an amount of silica far in excess of the metasilicate ratio.

b. The Solvent is CO_2.—Braune and Strassmann (6) studied the solubility of iodine in gaseous CO_2 at pressures up to 50 atm. and temperatures from 32.6 to 98.5°. The solubility effect of CO_2, expressed as the ratio of iodine actually present in the compressed gas to that which would be present if there were no solubility effect ranged up to almost three to one.

c. The Solvent is SO_2.—Niggli (51) demonstrated the solubility of $HgBr_2$ and HgI_2 in gaseous SO_2 by the increase in the critical temperature of the

TABLE 6

SOLUBILITIES IN SUPERHEATED STEAM CONTAINING ABOUT 7% CO_2 AT 500° C, 1,000 BARS, PARTS PER MILLION

	Without CO_2	With CO_2
$CaCO_3$	120	940
Fe_2O_3	80	230
NiO	20	43
SnO_2	2	50
Quartz	26,000	19,000

solvent. The critical temperature of SO_2 was found to be 157.6°. The solution at the first critical end point, 159.40, in the system SO_2—HgBr contained 1.5 wt. % $HgBr_2$; in the system SO_2—HgI, at 158.2°, 0.7%. His experiments were made in glass tubes, and it was not possible to realize the second critical end point, but in the region between the two end points, at 254°, in the system SO_2—HgI_2, a gas was obtained containing 6.2% HgI_2; in the system $HgBr_2$, 8.59. Pressures were not measured.

d. The Solvent is $H_2O + CO_2$.—Water containing dissolved CO_2 was used as a solvent in unpublished experiments by Morey, some of which are mentioned in the Annual Report of the Director of the Geophysical Laboratory (1), and these experiments are still under way. The apparatus and method used were similar to those used by Morey and Hesselgesser (38), and the method of obtaining a mixture of H_2O and CO_2 of constant composition was based on these two substances being immiscible in the liquid state. A stainless steel cylinder was filled with liquid CO_2, which under pressures greater than 7,500 psi is heavier than water. Then when water at 15,000 or 30,000 psi was pumped into the bottom of the cylinder it rose to the top and contained a little more than 7% CO_2. This mixture was passed over the material being studied in the same manner as was water

in the previous experiments. The same method of study could be used with H_2S or SO_2. The results with several substances are shown in Table 6. The effect of the carbon dioxide was to increase the solubility of tin oxide about 25-fold, of iron oxide about four-fold, and to decrease the solubility of silica about 20%. Decomposition was more evident than with water alone. The material carried in solution in the steam was almost all Na_2O and SiO_2, and the alumina accumulated in the residue.

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October 4, 1956

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APPARATUS AND TECHNIQUES FOR THE MEASUREMENT OF CERTAIN OPTICAL PROPERTIES OF ORE MINERALS IN REFLECTED LIGHT

EUGENE N. CAMERON

ABSTRACT

The paper discusses the measurement of rotation properties of anisotropic ore minerals in reflected light. The theory of measurement is first examined, and measurements according to the Berek and Hallimond methods are appraised. Three types of microscope equipment now available for quantitative work, including a new type of microscope produced by Bausch and Lomb Optical Company, are described, and procedures of measurement with each are given in detail.

INTRODUCTION

With the development of more sensitive and versatile microscopes, qualitative observations of ore minerals in reflected light can now be supplemented by quantitative determination of certain optical properties. The usefulness of optical properties in ore mineral identification is thereby greatly enhanced. The theoretical basis for quantitative work has been laid by Drude, Wright, Berek, Orcel, and others over a period of nearly eighty years, but the development of suitable apparatus has been slow. Most of the microscope equipment currently in use in various laboratories in this country is unusable for quantitative ore microscopy and gives only inconsistent and unreliable results. Several types of devices having the necessary optical quality and sensitivity are now available, however, and the purpose of this article is to describe them and discuss their usefulness in quantitative work. Among them is a new type of ore microscope designed and constructed by the Bausch and Lomb Optical Company in consultation with the writer.

ACKNOWLEDGMENTS

The writer is indebted to Mr. J. R. Benford, Bausch and Lomb Optical Co., for a critical review of the paper, with particular reference to the optical principles involved. Mr. John M. Guilbert and Mr. James F. Davis have also reviewed the paper and have made useful criticisms. The interest of Mr. Thomas Mitchell, Mr. Michael R. Seelinger, and Mr. D. J. Joerger of Bausch and Lomb Optical Company in quantitative work has greatly stimulated development and production of the new apparatus described herein. Their suggestions and assistance in many ways are gratefully acknowledged.

THEORY OF MEASUREMENT

Analysis of Elliptical Vibrations.—When two linear vibrations differing in phase and vibrating in directions at right angles to one another combine, the result is an elliptical vibration. This may be taken as the general case for light reflected from an anisotropic absorbing mineral having its two vibration directions at 45 degrees to the vibration plane of the polarizer. In Figure 1, the vibration directions of a mineral grain under observation are parallel to OE_1 and OE_2. A vibration OE parallel to OP, the plane of

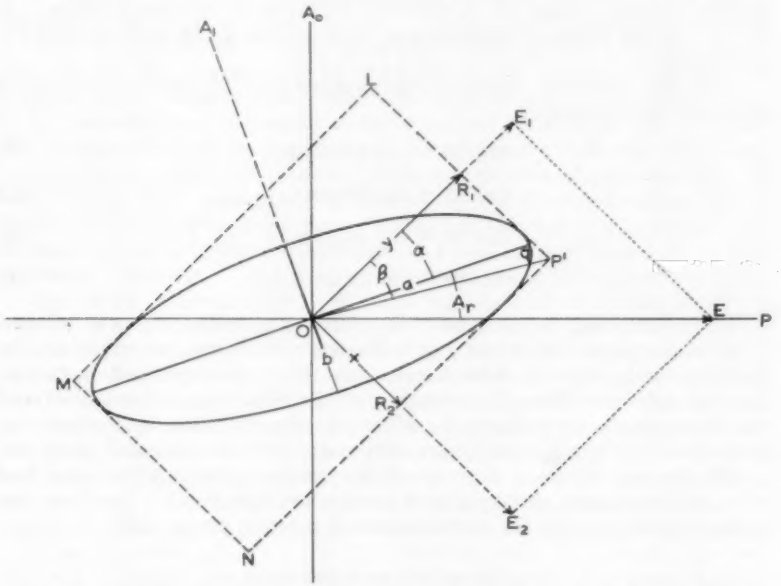

FIG. 1. Elliptical vibration produced by reflection of incident linear vibration OE by an anisotropic mineral at a 45° position. OE_1 and OE_2 are components of OE parallel to the vibration directions of the mineral; OR_1 and OR_2 are the corresponding reflected components. OP and OA_0 are planes of vibration of polarizer and analyzer (crossed position).

the polarizer, is resolved into two components OE_1 and OE_2. Part of each component is absorbed, part reflected. The two reflected components, differing in amplitude and phase, are $OR_1 = y$ and $OR_2 = x$. The ellipse of vibration of the ray produced by combination of OR_1 and OR_2 is always inscribed in the rectangle $LMNP'$, the half sides of which are defined by OR_1 and OR_2. The half diagonal OP' of this rectangle gives the vibration direction and amplitude of the linear vibration that would result from OR_1 and OR_2 if there were no phase difference between them. The semi-major and semi-minor axes of the ellipse of vibration are respectively a and b.

Referring to Figure 1, the following standard geometric relation holds:

$$x^2 + y^2 = (OP')^2 = a^2 + b^2. \tag{1}$$

Certain quantities must now be defined:

The ellipticity, $\vartheta = \tan^{-1} b/a$
Φ_x = the phase of $x(OR_2)$; Φ_y = the phase of $y(OR_1)$
$\Delta_{x,y} = \Phi_x - \Phi_y$ = the phase difference between x and y.

If $\Delta_{x,y}$ is > 0 the ellipse is left-handed; i.e., the motion of an ether particle during passage of an elliptical vibration is counterclockwise. If $\Delta_{x,y}$ is < 0 the ellipse is right-handed, and the motion of an ether particle is clockwise.

Now the following standard relationships hold for the ellipse and its circumscribed rectangle:

$$\tan 2\alpha = \tan 2\beta \cos \Delta_{x,y} \tag{2}$$

$$\cos 2\beta = \cos 2\vartheta \cos 2\alpha \tag{3}$$

$$\tan 2\vartheta = \tan \Delta_{x,y} \sin 2\alpha \tag{4}$$

$$\sin 2\vartheta = \sin 2\beta \sin \Delta_{x,y}. \tag{5}$$

In the foregoing, the analysis of the ellipse of vibration has been referred to the rectangle having x and y as half-sides. However, the ellipse can be similarly analyzed with reference to any other circumscribed rectangle. The half sides of each such rectangle give the directions and amplitudes of two components into which the elliptical vibration can be resolved by addition of an appropriate phase difference, and the diagonal gives the amplitude and vibration direction of the resulting linear vibration. For each such rectangle, the equations given above are valid. They are the mathematical basis for the measurement of rotation properties.

METHODS OF MEASUREMENT

General Statement.—From equations (2)–(5) given above, it will be evident that if any two of the quantities $\Delta_{x,y}$, α, β, and ϑ are known for any ellipse and a given circumscribed rectangle, the other quantities can be calculated and the elliptical vibration thereby defined. Furthermore, if the quantities are known for an ellipse and one circumscribed rectangle, mathematical relationships between the ellipse and any other circumscribed rectangle of known orientation can be calculated.

Various methods of measurement differ basically according to the orientation of the circumscribed rectangle used for analysis or according to the specific pair of quantities measured. For a full review of various methods, reference may be made to Berek (2), Königsberger (7), Wright (10), Orcel (8), Turner and others (9), Cameron and Green (3), and Hallimond (5). The present article discusses two methods appropriate to available equipment. A third method is briefly mentioned.

In the first two methods, here designated the Hallimond and Berek methods, the analyzer is used for measuring an angle or angles of rotation, and a thin rotatable mica plate is used to compensate the phase difference between two components of the elliptical vibration produced by the mineral. Scales and verniers permitting settings of the analyzer and mica compensator to 0.1° must be available. In each method, it is necessary first to determine the zero settings of the analyzer and compensator. The zero setting for the analyzer is the position at which it is crossed with the polarizer. This can be determined by examining the polarization figure of an isotropic mineral. At the zero position, with the mica plate out of the system, the figure is a perfect cross. The zero position of the mica plate can now be determined. If the mica plate is introduced into the system and rotated, the zero position of the plate is that position at which the blackest perfect cross is seen. When the mica plate is at its zero position, its fast ray direction is parallel to the vibration direction of the analyzer. The zero positions of the analyzer and compensator are recorded for reference in measurement of rotation properties of anisotropic minerals. If both settings have been properly determined and if other possible defects in the optical system have been eliminated, the figure of any uniaxial mineral will be a perfect black cross at 4 positions 90° apart in a 360° rotation of the stage. All measurements are made in monochromatic light.

Hallimond Method.—The most practical method of measurement of rotation properties is that described by Hallimond (5, p. 122–130). An anisotropic mineral (polished surface) is placed on the stage, the stage is rotated with analyzer and polarizer crossed, and the four extinction positions of the mineral are determined. If these fall at 90° from one another, then the polars are exactly crossed. The mineral is then rotated to a position at 45° to one pair of adjacent extinction positions. If the extinction positions are not 90° apart, the polars are not precisely crossed. (It is assumed here that the optical system is free of defects, such as strain in objectives, that would produce the same effects on extinction positions as uncrossing of the polars.) The 45° positions then lie halfway between adjacent pairs of extinction positions. For example, if a given mineral shows extinction at the 40°, 120°, 220° and 300° settings of the stage, the 45° positions are at 80°, 170°, 260°, and 350°.

The stage is now rotated to a 45° position, and the analyzer is rotated until extinction at the middle of the field is produced. This can be determined conoscopically by observing the polarization figure. At the 45° position the figure consists of two separated isogyres. Rotation of the analyzer either to right or left, depending on the direction of rotation of the incident ray by the mineral, causes the isogyres to approach one another and finally to form a cross. The analyzer is now at A_1 perpendicular to $a = Oq$ (Fig. 1). The angle of rotation of the analyzer from A_0 to A_1 is the angle of apparent rotation (A_r) of the mineral, i.e., the angle $POq = 45° - \alpha$. Thus far the method is the same as that employed by the writer and others (3, 4), and the reader is referred to the earlier articles for details of the procedure and precautions to be taken. If the two rays

reflected from the mineral differ appreciably in phase, however, true extinction is not produced at the center of the field, hence the center of the cross is partly illuminated. The correct position of A_1 ($\perp Oq = a$) is now a position of minimum illumination, rather than extinction. The greater the phase difference the less complete the extinction and the more difficult is the precise setting of the analyzer. To overcome this difficulty, Hallimond has introduced the use of the Nakamura plate (5, p. 75, 123–138).

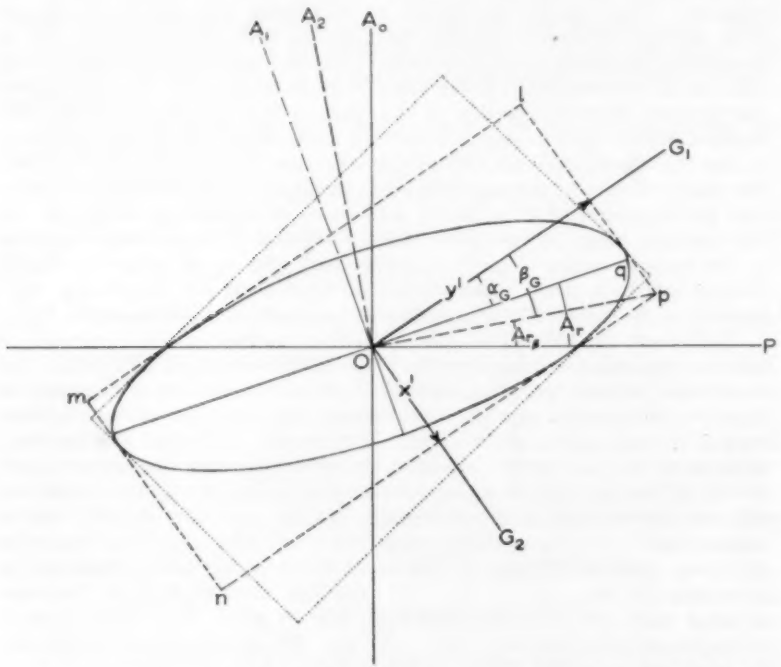

FIG. 2. Diagram to show relationships at compensation, for a right ellipse of vibration. The dotted rectangle is rectangle $LMNP'$ of Figure 1. P—vibration direction of polarizer; A_σ—vibration plane of analyzer at crossed position. OG_1 and OG_2—slow and fast ray directions of mica compensator.

This is a thin biquartz plate that is sensitive to small rotations. The plate is inserted in the system in such a way that the boundary between the two halves of the plate passes through the center of the field and can be viewed by means of an ocular. The slightest deviation (commonly less than 0.1°) of the analyzer from the correct setting produces a contrast in the illumination of the two halves of the plate. Except with a few minerals that produce unusually large phase differences, the Nakamura plate permits readings of the angle of rotation with a maximum error of ± 0.2°.

The analyzer is left in the system, and the mica plate is now inserted and rotated until maximum darkness is produced under orthoscopic observation, or better, until the center of the polarization cross as seen conoscopically is made as black as possible. Hallimond recommends the use of a Königsberger plate for determining the exact setting of the compensator. This is a very thin bimica plate with two halves that are of equal thickness but are oriented so that fast and slow rays in one half are at 90° to those in the other half. The plate is sensitive to small phase differences but insensitive to small rotations. The device is unfortunately not available at present, due to difficulties in manufacture.

The action of the mica compensator can be followed by means of Figure 2. On entering the mica plate, the elliptical vibration due to the mineral is resolved into two component linear vibrations, x' and y', differing in phase. The two components are parallel respectively to the vibration directions OG_1 and OG_2 of the mica plate. OG_1 is the slow ray direction. The components x' and y' now define the half sides of a new rectangle $lmnp$ circumscribed about the ellipse. In passing through the mica plate x' and y' will acquire an additional phase difference that is dependent on the optical thickness of the mica plate. At compensation, the phase difference produced by the mica plate must be equal and opposite to the initial phase difference between x' and y'. In order that this condition may be satisfied, the following must hold for the relation of the ellipse of vibration of the mineral to the circumscribed rectangle defined by the mica plate vibration directions:

If Δ_G = the phase difference between slow and fast rays of the mica plate, and $\Delta_{x',y'}$ = the phase difference for the two components of the mineral ellipse of vibration parallel to OG_1 and OG_2, then at compensation, $\Delta_G = \Delta_{x',y'}$, and from (4)

$$\tan \Delta_G \sin 2\alpha_G = \tan 2\vartheta \tag{6}$$

The meaning of 2ϑ must now be analyzed. Let us suppose that a mica plate or other compensator is oriented with its vibration directions OG_1 and OG_2 at 45° to the major axis of a reflected ellipse of vibration (Fig. 3). On entering the compensator, the ellipse will be resolved into the two components x'' and y'' at 45° to the major axis of the ellipse. At the moment of entrance, the two components will have a phase difference due to the mineral. If this is matched by an equal and opposite retardation on passage of the components through the compensator, then on exit from the compensator the two components will be equal in phase and hence will combine to produce a linear vibration which will be Op coinciding with Oq. Hence in Figure 3, $2\alpha_G = 2\beta_G = 90°$, and in Eq. (6) $\tan \Delta_G = \tan 2\vartheta$. Thus 2ϑ for any elliptical vibration equals the phase difference between the components of the ellipse of vibration at 45 degrees to the major axis. These two components are the half sides of the rectangle with diagonal coinciding with the major axis of the ellipse.

The Hallimond method therefore consists of determining the azimuth ($A_r = A_0 - A_1$) of the major axis of the ellipse of vibration (Fig. 2) re-

ferred to the vibration plane of the polarizer, together with the azimuth (α_G) of the slow ray of a mica plate set at the compensation position. The angle α_G together with the known phase difference of the mica plate is used to calculate 2ϑ, the phase difference for the two components at 45° to the ellipse, from Eq. (6). In practice, A_r and α_G are each measured at two adjacent 45° positions. The mean A_r and α_G are taken as the final values and used in calculating 2ϑ.

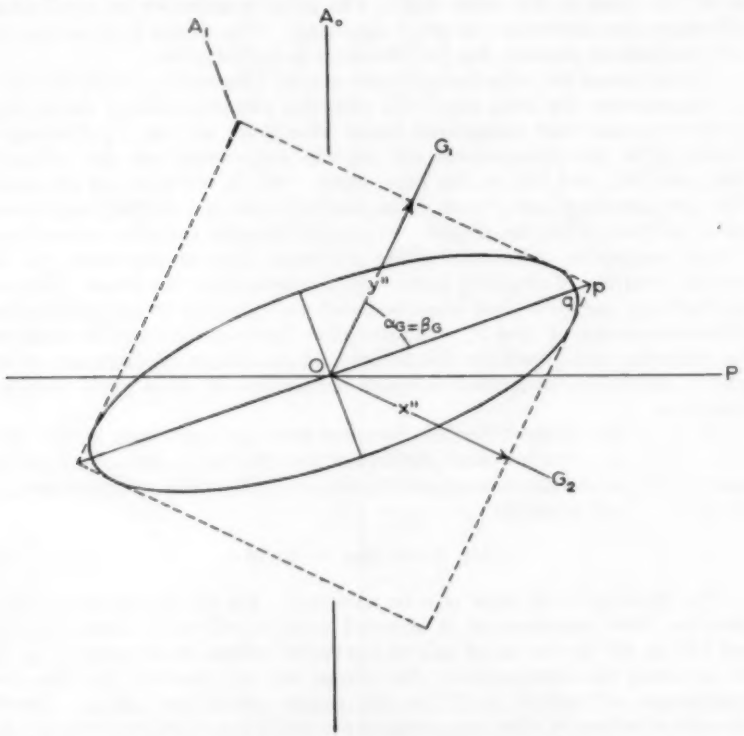

FIG. 3. Diagram showing relations at compensation, where $\Delta_G = \Delta_{x'',y''}$.

Berek Method.—It has been shown that when the phase difference produced by the mineral is compensated by rotating the mica compensator to the correct position, two components that are parallel to OG_1 and OG_2 and that are equal in phase emerge from the mica plate. Upon emergence, these will combine into a linear vibration having the direction Op (Fig. 2), which will not be parallel to Oq. In order to produce true extinction, the analyzer must now be rotated by an amount equal to the angle qOp, to the position A_2 perpendicular to Op. The difference between this new setting and the crossed position of the analyzer A_0 is the angle $45° - \beta$.

In practice, measurement by the Berek method starts with the mineral at a 45-degree position and the analyzer and mica compensator both at their zero positions. The two are then simultaneously rotated back and forth until the position of extinction is found. The weakness of this method is that accurate setting of the analyzer is dependent upon accurate setting of the mica plate, and this, depending on the size of the phase difference produced by the mineral and the specific retardation of the mica plate, is often difficult or impossible to do. The error in setting of the mica plate may be as much as ± 4 degrees when $\Delta_{x,y}$ is appreciable, and errors therefore arise in setting the analyzer. In addition, the exact value of angle *POp* depends on the phase difference of the mica plate used, and no two plates will have precisely the same phase difference. In order to convert measurements to standard values, cumbersome calculations using equations (2) to (5) are necessary. The Hallimond method is therefore much the preferable one for general use. It allows precise measurement of angle A_r, and this precision is not affected by any subsequent error in setting the mica plate. The apparent angle of rotation measured is independent of the mica plate thickness, and a standard maximum value for any mineral can be established for any given wavelength of light. This value can be reproduced by any observer.

The difference between the Hallimond rotation angle (A_r, Fig. 2) and the Berek rotation angle *POp* or $A_{r\vartheta}$, (Fig. 2) can be calculated from equations (2) to (5). For example, let us suppose that for a given right ellipse:

$$\Delta_{x,y} = 20^\circ \text{ and}$$
$$\beta = 40^\circ; \quad \text{hence } 2\beta = 80^\circ$$
$$\text{Then } \tan 2\alpha = \tan 80^\circ \cos 20^\circ \quad \text{from (2)}$$
$$\tan 2\alpha = 5.3292$$
$$2\alpha = 79^\circ\, 22'\, 20''$$
$$\text{Next, } \tan 20^\circ \sin 79^\circ\, 22'\, 20'' = \tan 2\vartheta \quad \text{from (4)}$$
$$\tan 2\vartheta = 0.35775$$
$$2\vartheta = 19^\circ\, 41' = 19.68^\circ.$$

This is the phase difference for the components at 45° to the ellipse.

Using Figure 2 as a general guide, the above values may be used to calculate the setting of the mica compensator over the ellipse, hence angle α_G.

$$\text{let } \Delta_G = 36^\circ \left(\frac{\lambda}{10}\right) = \text{the phase difference for the compensator}$$

$$\text{Then } \tan \Delta_G \sin 2\alpha_G = \tan 2\vartheta \quad \text{from (4)}$$

$$\sin 2\alpha_G = \frac{\tan 19^\circ\, 41'}{\tan 36^\circ} = \frac{0.35775}{0.72654} = 0.49242$$

$$2\alpha_G = 29^\circ\, 30'\, 00''$$
$$\alpha_G = 14^\circ\, 45' = 14.75^\circ.$$

The azimuth β_G (Fig. 2) of the linear vibration produced by compensation in the mica plate must next be determined.

$$\tan 2\alpha_G = \tan 2\beta_G \cos \Delta_G \quad \text{from (2)}$$

$$\tan 2\beta_G = \frac{\tan 29^\circ\ 30'\ 00''}{\cos 36^\circ} = \frac{0.5658}{0.80902} = 0.6994$$

$$2\beta_G = 34^\circ\ 58'$$

$$\beta_G = 17^\circ\ 29' = 17.48^\circ.$$

The difference between the rotation angles measured by the Berek and Hallimond methods has been shown to equal the angle qOp, or $\beta_G - \alpha_G$. This angle equals 2.73° under the conditions assumed above.

TABLE I

DIFFERENCES BETWEEN HALLIMOND AND BEREK ROTATION ANGLES

$\Delta_{x,y}$	$\beta = \left(\tan^{-1}\frac{OR_2}{OR_1}\right)$	α_G	β_G	$\beta_G - \alpha_G$
5°	35°	3.2	4.0	0.8
10°	35°	6.6	8.1	1.5
15°	35°	10.1	12.2	2.1
20°	35°	13.7	16.6	2.9
25°	35°	18.3	21.3	3.0
30°	35°	23.6	26.5	2.9
5°	40°	3.4	4.2	0.8
10°	40°	6.9	8.5	1.6
15°	40°	10.6	12.8	2.2
20°	40°	14.7	17.5	2.8
25°	40°	19.5	22.5	3.0
30°	40°	25.6	28.5	2.9
5°	43.5°	3.5	4.3	0.8
10°	43.5°	7.0	8.7	1.7
15°	43.5°	11.2	13.5	2.3
20°	43.5°	15.0	17.8	2.8
25°	43.5°	19.9	23.0	3.1
30°	43.5°	26.2	29.0	2.8

In general, the difference between β_G and α_G is a function of $\Delta_{x,y}$ and $\frac{OR_1}{OR_2}$ of the mineral, and Δ_G for the mica plate used. Table I shows variations in $\beta_G - \alpha_G$ with $\Delta_{x,y}$ and $\frac{OR_1}{OR_2}$ for a mica plate with $\Delta_G = 36°$. The data are calculated by the method exemplified above. Similar calculations can be made to show the variation in $\beta_G - \alpha_G$ with Δ_G. For example, if $\Delta_{x,y} = 15°$, $\beta = 40°$, and $\Delta_G = 36°$, then as shown in the table $\beta_G - \alpha_G = 2.2°$. But if $\Delta_G = 30°$, $\beta_G - \alpha_G = 1.75°$. For very small phase differences, $\beta_G - \alpha_G$ is negligible relative to the accuracy of measurement ($\pm$ 0.2° to $\pm$ 0.3°). $\beta_G - \alpha_G$ also becomes negligible as the phase difference for the mineral approaches the phase difference for the mica plate.

Relationships shown in Figure 2 and used as a guide to the above calcu-

lations are for a right ellipse ($\Delta_{x,y} < 0$). Note that angle α_G is the angle between the slow ray direction of the mica plate and Oq, which is perpendicular to A_1. The total rotation of the mica plate from its zero position must therefore be corrected for the rotation of the analyzer from A_0 to $A_1 = A_r$. For example, let us suppose that the zero position of the mica plate is at 45° on the scale. At compensation, the setting of the mica plate is at 75°. Then in Figure 2,

$$\alpha_G = (\text{total rotation of mica plate}) - A_r = G_1OP - A_r$$
$$\alpha_G = (75° - 45°) - A_r = 30° - A_r.$$

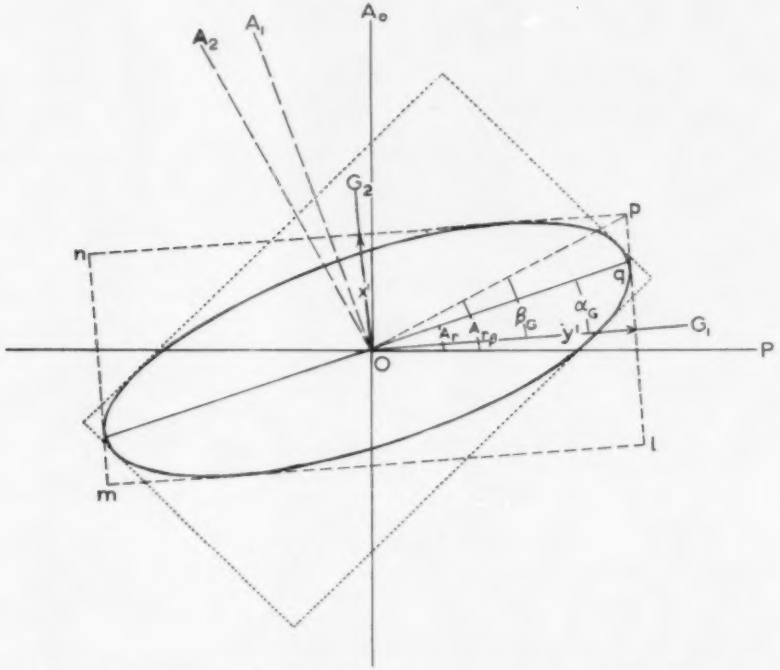

FIG. 4. Diagram to show relations at compensation, for a left ellipse. P—vibration plane of polarizer; A_0—vibration plane of analyzer at crossed position. OG_1—slow ray vibration direction of mica compensator; OG_2—fast ray vibration direction. The dotted rectangle corresponds to rectangle $LMNP'$ of Figure 1.

The sign of α_G in this case is to be taken as negative. Relations for a left ellipse ($\Delta_{x,y} > 0$) are shown in Figure 4. Here $\alpha_G = A_0OA_1 - (\text{total rotation of the plate})$.

$$\alpha_G = A_0OA_1 - AOG_2$$
$$\alpha_G = POq - POG_1.$$

The sign of α_G in this case is to be taken as positive. For a given mineral, the sign of α_G may be the same for all wavelengths, or it may change sign with wavelength, passing through zero at some intermediate wavelength.

MacCullagh Method.—In the last edition of the work of Rinne and Berek (2, p. 323) a method ascribed to J. MacCullagh is recommended for the analysis of rotation properties. An analysis of the relations involved was given by Horn (6). If a rotating mica plate having a phase difference between 2ϑ and $180 - 2\vartheta$ for the mineral under observation is used as a compensator, there are two positions at which the plate compensates the phase difference produced by the mineral. One is at the azimuth of OG_1 defined by angle α_G in Figure 2, the other is defined by the angle $90° - \alpha_G$. Rinne and Berek recommend using measurements of the Berek rotation angle and angle α_G (Fig. 2) at the two positions of compensation for calculating the other quantities defining an ellipse of vibration. Unfortunately, even apart from the potential errors due to inaccurate setting of the mica plate, the method is mechanically unworkable with any microscope apparatus now available, including the Berek ocular, unless $\Delta_{z,y}$ closely approaches Δ_G.

APPARATUS FOR MEASUREMENT OF ROTATION PROPERTIES

General Description.—Three types of apparatus are now available for quantitative measurements in reflected light with the upright type of microscope. One is the Berek ocular, made by E. Leitz. The second is a combination of slotted Wright ocular and elliptical compensator made by Cooke, Troughton and Simms, Ltd. The third is a new ore microscope made by Bausch and Lomb Optical Company. The three types of apparatus differ markedly in design.

Berek Ocular.—This ocular (Fig. 5), designed and described by the late M. Berek, was the first modern device produced for quantitative work of the kind under discussion. It is used in place of the regular ocular. The device includes an analyzer that can be rotated by means of a knob through an angle of 90°, the zero position being located at the middle of the scale. Beneath this is a similarly rotatable mica plate. When the plate is at zero position, it is in the middle of its scale and has its slow ray normal to the vibration plane of the analyzer when the latter is at zero position. Verniers on both analyzer and mica plate scales allow readings to 0.1°. A focussing auxiliary lens can be swung over the ocular when conoscopic examination is desired.

The ocular is intended for measurements by the Berek method. Its only disadvantage for this purpose is that the ocular cannot be removed and a pinhole ocular inserted when a small bright polarization figure is desired, as is the case for some minerals. If the phase difference produced by the mineral under observation is small it can be used for measurements by the Hallimond method. If the phase difference is enough to require use of the Nakamura plate, however, difficulties arise. The Nakamura plate must be placed in the tube slot, the swing-out auxiliary lens must be

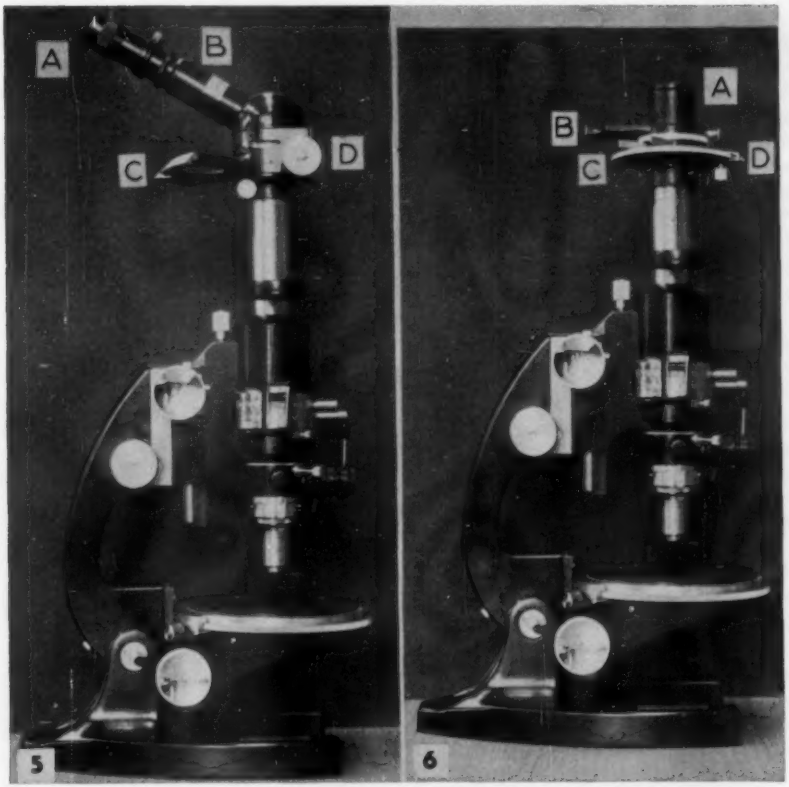

FIG. 5. Berek ocular (E. Leitz) mounted on microscope in place of the usual ocular. *A*—auxiliary lens (swung into place over ocular) to permit conoscopic observation. *B*—focussing ocular. *C*—arc with scales and verniers for reading positions of analyzer and elliptical compensator. *D*—turning knob for rotating analyzer; a similar knob on the opposite side of the device controls rotation of mica plate.

FIG. 6. Slotted Wright ocular (Cooke, Troughton, and Simms) mounted in place of the usual ocular. *A*—eyepiece. *B*—sliding analyzer. *C*—circle divided on upper surface into 360°. *D*—vernier. Slot for elliptical compensator or Nakamura plate is between analyzer and divided circle.

used to focus on the plate boundary, and the mica plate rotated to the position at which the two halves of the plate at the center of the figure are equal in hue and intensity. Unfortunately, at the focal point, the plate boundary is seen superimposed over the polarization figure, and the correct setting is very difficult to determine.

The Slotted Wright Ocular.—This apparatus is manufactured by Cooke, Troughton and Simms, Ltd. Its use in connection with a mica com-

pensator and Nakamura plate has been discussed in detail by A. F. Hallimond (5). The device (Fig. 6) is substituted for the usual microscope ocular. It consists of an ocular beneath which a sliding analyzer is mounted. Beneath the analyzer is a slot into which an elliptical compensator can be inserted. The ocular with its slots can be turned through 360° and the setting on a circular scale read to 0.1° by means of a vernier. The elliptical compensator consists of a mica plate which can be rotated through an angle of about 100°. The zero position of the plate is near the middle of its scale. Scale and vernier on the plate permit reading the setting to 0.1°. The slot in the ocular also will accommodate the Nakamura plate, the boundary line of which can be focussed by sliding the ocular in its mount. In this case the compensator is used in the tube slot of the microscope. Observation may be done either orthoscopically or conoscopically, the latter by removing the lens or by use of an auxiliary lens. In our laboratory we have found it best to determine the rotation angle orthoscopically with the Nakamura plate in the ocular slot. The Nakamura plate is then withdrawn, the elliptical compensator is inserted, and the polarization figure is used for setting the mica compensator at the correct position. Hallimond has suggested use of the Nakamura plate in the tube slots where its image is superimposed on the polarization figure, but we have not found this satisfactory.

Good results can be obtained by use of the apparatus described above. Its only drawback is that it is somewhat inconvenient to use, because in moving from observation to reading the scales the eye must constantly adjust focus.

Bausch and Lomb Microscope.—A new Bausch and Lomb apparatus, a modification of the standard Model O ore microscope, is especially well suited for measurement of rotation properties by its simplicity of design (Fig. 7) and convenience of use. It is designed for precise work, yet the special optical components introduced in no way interfere with or complicate use of the microscope for the most ordinary observations. The polarizer is pinned in place in the horizontal illuminator tube. Its orientation can be adjusted by loosening the lock nut on the collar of the tube and rotating the tube. The reflecting plate in the tube of the microscope is of the coated type. The analyzer is a polaroid plate which is rotatable by means of a handle, whereby it can also be inserted and withdrawn. The analyzer is provided with a scale and vernier by means of which it can be set to 0.1°. Beneath the analyzer is mounted a mica plate which can be rotated, inserted, and withdrawn by the same type of handle as the handle of the analyzer. A scale and vernier permit setting the mica plate to the nearest 0.1°. The analyzer and mica plate settings are read simultaneously through a right-angled reflecting prism. This is viewed from above by means of a lens that can be focussed on the scales. The lens is slid up and down until a position is found at which the eye requires no adjustment of focus in moving from observation of the mineral to reading of the scales. This device contributes greatly to the ease and speed of observation and measurement.

Measurements may be made conoscopically or orthoscopically, according

to whether the mineral produces an appreciable phase difference or not. A 6 mm objective is the most satisfactory unless excessively small grains (less than .05 mm) are to be observed. When a grain showing maximum anisotropy has been selected for measurement, the 45° positions of the

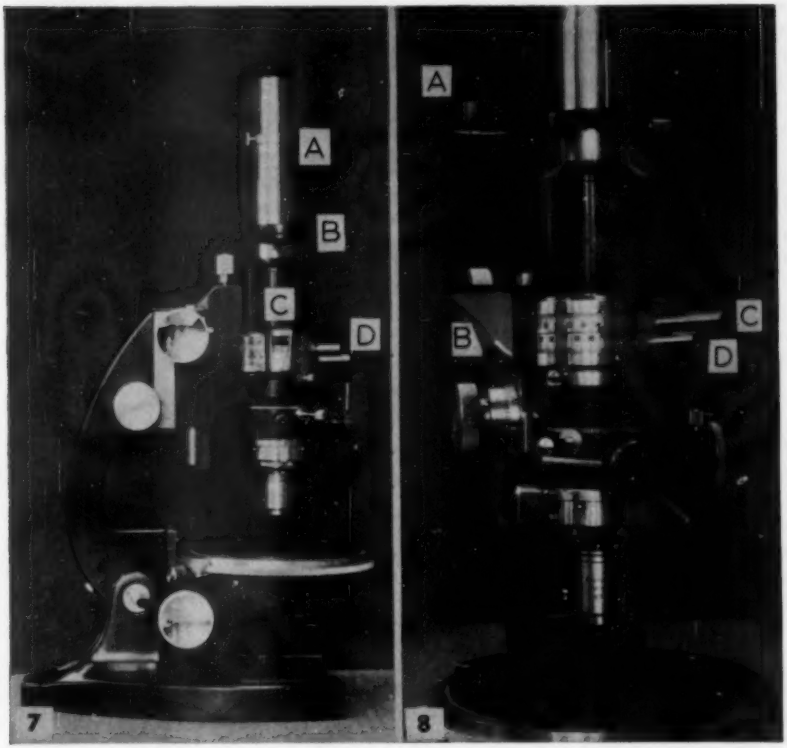

FIG. 7. Bausch and Lomb ore microscope. *A*—focussing phase telescope. *B*—viewing lens for reading scales through right prism *C*. *D*—analyzer handle (upper) and mica compensator handle (lower). Scales rotate as handles are moved.

FIG. 8. Bausch and Lomb microscope, showing details of compensating devices. *A*—sliding, focussing viewing lens, for reading scales through prism *B*. *C*—handle of analyzer. *D*—handle of elliptical compensator.

mineral are found and the stage is rotated to one of them. If the mineral is one giving a small phase difference, conoscopic measurement is the simplest procedure. The ocular is removed and the polarization figure is used for measurement. Angle A_r is determined by means of the analyzer. The mica plate is then inserted and angle α_G is determined by compensation of

the center of the figure. The stage is turned to the next 45° position and the measurements are repeated. The two sets of readings are averaged.

If the mineral shows an appreciable phase difference, and whenever especially precise results are desired, orthoscopic observation and the use of the Nakamura plate are necessary. The mineral is set as before to a 45° position and the objective centered. The usual ocular is replaced by a special focussing phase telescope, and the mineral is brought into focus by racking the stage downward from the focal position for the normal ocular. The Nakamura plate is inserted in the tube slot, the telescope is focussed upon it by moving the sliding eye lens, and the boundary between its two halves is centered by moving the plate until the boundary passes through the optical center of the field. The field now appears as two illuminated semicircles that are separated by the plate boundary and appear superimposed on the image of the mineral. The analyzer is turned until at the center of the field the two sides of the plate are equal in color and intensity of illumination. A_r is then calculated from the setting of the analyzer $(A_0 - A_1)$. The Nakamura plate and ocular are now removed, the mica plate is inserted, the stage racked up until the polarization figure appears, and angle α_G is determined. The measurements are repeated at adjacent 45° positions.

CORRECTION OF RESULTS

As discussed by the writer and others (4), the values of A_r determined by any of the methods described above must be corrected for rotation during upward passage through the reflecting plate. The values of α_G must also be corrected for passage through the plate. The correction of α_G is very small, except for minerals of high rotation angles ($> 5°$), and Hallimond points out (5, p. 127) that the same correction factor can be used for both the rotation and ellipticity. The corrections will always be reductions of the observed values.

MEASUREMENT OF DISPERSION

The dispersion of a mineral is an important property for determinative purposes. It is determined by measuring A_r and α_G in various wavelengths of light, and calculating 2ϑ from the measurements of α_G. The value of Δ_G must be known for each wavelength of light. This information should be furnished by the manufacturer.

OTHER EQUIPMENT

For quantitative work, properly centered aperture and field diaphragms, a coated reflecting plate, and a well centered strong source of illumination must be provided. For observations with polarization figures, strain-free 4-mm and 6-mm objectives are necessary. Such objectives can be provided by any optical manufacturer by employing careful selection. It is not necessary that the objectives be completely free of strain, but only that the strain in the center of the objective lenses be no more than enough to cause the center of the polarization figure to become gray as the objective is

rotated in its collar. The figure should not break into isogyres. If a strain-free objective develops strain with age, it should be returned to the manufacturer for resetting of the components.

The Nakamura plate is insensitive to small phase differences (5, p. 128); hence a strained objective, even a 16 mm objective, can be used in measurement of A_r. The objective should be set at one of its extinction positions. To do this, the crossed position of the analyzer and polarizer is found, using the figure of an isotropic mineral and a strain-free objective. The strained objective is now substituted for the other and rotated in its collar to a position at which the polarization figure is a perfect cross. It may now be used for measurements of A_r with the Nakamura plate. A 16 mm objective has the advantage that it gives a brighter image than an objective of higher magnification.

For oil immersion work, a 40 ×, N.A. 1.00 Bausch and Lomb fluorite objective is the only one we have found suitable for use with the Nakamura plate. The 90 × to 95 ×, N.A. 1.25 objectives in our laboratory all permit too much stray light, so that contrast in the Nakamura plate is obscured. There are probably other oil immersion objectives not available in our laboratory, however, that will prove suitable.

For work in monochromatic light, the ordinary low-voltage tungsten lamps provide light enough only in the range from about 520 mμ to 600 mμ. The light even in this range is adequate only if a variable transformer is used and the voltage on a 6-volt lamp increased to 10 or 12 volts. If in studying a mineral this voltage is maintained only during actual measurements, the life of the average bulb is not significantly shortened. For monochromatic light, however, cap filters are necessary with built-in light sources. This is a nuisance and will remain so until filters easily inserted into the optical system are available. For precise work in monochromatic light over the range 450 to 650 mμ, a carbon arc lamp is still the only recourse. A microscope with a stage that can be racked up and down is therefore essential.

GENERAL COMMENT

The obstacles to quantitative measurement of optical properties of ore minerals have been removed to such an extent that this work is now brought within the scope of routine investigation of ore minerals. Improvements in apparatus will undoubtedly be made from time to time, but the necessary optical elements are already available. The need now is for measurements of the rotation properties of the various opaque and semi-opaque mineral species. This work is in progress in our laboratory, and data will be published from time to time as they accumulate. Those who attempt to make use of quantitative techniques, however, should not expect to obtain consistent or reliable results with equipment that was designed and manufactured only with qualitative observations in mind.

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MADISON, WIS.,
Dec. 1, 1956

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SULFUR ISOTOPES AND MINERAL PARAGENESIS[1]

M. L. JENSEN

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ABSTRACT

Almost all of the previous investigations of S^{32}/S^{34} ratios of sulfide minerals and ores have indicated a lack of significant variations in the ratios of specimens collected from one deposit. Most of these valuable previous studies have been done by non-geologists upon, unfortunately, limited and poorly representative mineral suites of sulfide deposits with little regard for the geologic occurrence and paragenesis of the sulfides present in the given deposit.

This study of S^{32}/S^{34} ratios obtained from two or more sulfides occurring in one hand specimen indicates that appreciable variations, greater than four percent in one example, are evident. Therefore, a partial attempt is made to analyze the significance of variations in S^{32}/S^{34} ratios by means of a more detailed study of the genesis and the paragenetic relationships of the various sulfides present in selected specimens.

The isotopic and geologic study of the selected specimens suggests the following: (1) a suggestion for distinguishing between exsolution and replacement is offered by isotopic ratios, (2) the mechanism of replacement of one sulfide by another is not always a diffusion mechanism of the small metal cations but also includes an exchange of the large sulfur anions, (3) hypogene ore fluids, giving rise to a vein, may vary in isotopic composition as indicated by an isotopic study of the minerals formed, and (4) the isotopic reactions that take place during supergene enrichment suggest that supergene and hypogene minerals can be distinguished by isotopic analyses. Other suggestions and avenues of investigation are also discussed.

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INTRODUCTION

In a previous preliminary paper (9), I suggested, through the study of stable isotopic ratios, the possibility of distinguishing between supergene and hypogene minerals and, also, the possibility of fractionation of stable isotopes occurring during the primary hypogene stage of sequential formation or successive deposition of minerals. S^{32}/S^{34} investigations, however, both previous and subsequent to my suggestions, by Thode and coworkers (13, 14, 15, 18, 19, 20, 21) and by Kulp et al. (10), have shown no conclusive evidence of significant variations in sulfur ratios of sulfide minerals collected from one mineral deposit, nor in many cases, variations in sulfides collected from mining districts. The enigmatic Franklin Furnace district and the Mississippi Valley district are two exceptions.

This paper has a two-fold purpose. First, to call attention to the fact that appreciable variations in the S^{32}/S^{34} ratio of sulfide minerals, not only in one mineral deposit but *in one hand specimen*, have been measured and do exist—variations greater than 4 percent. Second, to illustrate that these variations in one mineral deposit and even in one hand specimen have a greater significance when studied in corroboration with a knowledge of the geologic occurrence and paragenesis of the various sulfides present in the deposit or in the hand specimen.

Quite frankly, at this stage, few facts are known about the specific geologic processes that caused these sulfide minerals to be formed with significantly different S^{32}/S^{34} ratios. Nevertheless, enough is known to allow certain suggestions pertaining to the mechanisms of both the mineral paragenesis and the enigmatic process of replacement, suggestions based upon the results of isotopic studies.

EXPERIMENTAL PROCEDURE

All but one of the sulfide samples analyzed were selected from the collections at Yale University. At first, specimens containing two or more sulfides exhibiting what I believe to be fairly evident paragenetic relationships were selected, but as the study continued sulfides of more complex and inconclusive paragenetic relationships were analyzed. Isotopic analyses were made only of those specimens where the respective sulfide phases could be separated mechanically from each other, with very little or no intermixing.

The sulfur dioxide gas was prepared by a standard procedure briefly described previously (2) and modified after a method described by Thode et al. (19), in which a stream of oxygen passes over the heated sulfides. One tank of pre-purified oxygen was used as a standard source for all samples and the flowage and burning rates were controlled by means of a bubble counter, manometers, and time schedules. The SO_2 formed was collected in a liquid nitrogen trap, transferred to a sample flask, and analyzed on a model 21-401 Consolidated Electrodynamics Corporation mass spectrometer.

For purposes of comparison with other published S^{32}/S^{34} studies, all ratios are related to the common standard of troilite from the Cañon Diablo meteorite with a ratio of 22.21, and the precision of the results is $\pm$ 0.01 in standard deviation or better unless otherwise indicated.

TABLE 1

DESCRIPTION OF SPECIMENS

Specimen	Cat. Number	Location	Mineralogic Description	S^{32}/S^{34}
A	— (Specimen obtained from Mr. D. J. Milton, Harvard Univ.)	Mackinaw Mine, Snohomish County, Washington	Nickel-gold ore specimen consisting of aggregate of coarse grained sulfides. Contains abundance of chalcopyrite and cubanite showing rather definite exsolved textures, e.g., inclusions of blades and rectangular phases having sharp smooth boundaries of cubanite in chalcopyrite, lack of consistent continuous orientation of blades in adjacent chalcopyrite-cubanite grains, and contraction in width of blades at mutual intersections.	
			Cubanite	22.10
			Chalcopyrite	22.10[4]
B (Fig. 1)	XVI-D12 Sud 139	Frood Mine, New deposit w. sect. 2600 stopes, Sudbury, Ontario	Relatively coarse bands, parallel to hanging wall, several millimeters thick consisting of pyrrhotite and chalcopyrite.	
			Bands of chalcopyrite	22.18[5]
			Bands of pyrrhotite	22.19
C	Brush 816	Bristol, Conn.	Veinlets of bornite and minor chalcopyrite cutting and replacing calcite. Chalcopyrite appears to have replaced bornite.	
			Bornite	22.27
			Chalcopyrite	22.28
D	Brush 1099	Bristol, Conn.	Typical aggregate of orthorhombic crystals of chalcocite, for which Bristol is well known.	22.9 ±0.1
E (Fig. 2)	—	Ikuno mine, Japan	Cockade ore specimen. Cores consist of earlier brecciated gangue and ore fragments, followed by successive deposition of quartz, sphalerite, and pyrite, and filling of remaining voids with calcite.	
			Early sphalerite crust	22.25[6]
			Later pyrite crust	22.26
F (Fig. 3)	Brush 699	Marsden's mine, Galena, Ill.	"Stalactite" consisting of a core of an unaltered euhedral galena crystal crustified with coating of ~1–3 mm dia. marcasite crystals. No evidence of oxidation on surface of galena crystal, therefore presumably formed entirely by hypogene fluids.	
			Galena	22.09
			Marcasite	22.39[1]
G (Fig. 4)	Brush 933	Vigsnus, Norway	Massive fine-grained pyrite forms base upon which grew large (1–2 cm dia.) crystals of sphalerite.	
			Pyrite	22.11
			Sphalerite	22.11
H (Fig. 5)	V-5	Unknown	Crustified vein specimen consisting of successive 1–3 cm thick layers, extending from wall rock to interior of vein, of (1) quartz, (2) galena, (3) quartz, (4) sphalerite, (5) galena, and (6) mixture of sparse, very fine-grained sulfides in jasper.	
			(2) Galena	22.39
			(4) Sphalerite	22.21
			(5) Galena	22.19
I (Fig. 6)	XII-D9 Kn 64	Kennecott, Alaska	Approximately 3 cm wide very coarse-grained veinlet of covellite with minor digenite remaining on two sides as enclosing host mineral. The covellite veinlet is cut and replaced by relatively smaller veinlets, some of which are composed of a fine-grained mixture of covellite and chalcocite (digenite?), whereas others consist entirely of digenite.	
			Enclosing digenite host	22.64
			Coarse grained covellite veinlet	21.91
			Small veinlet consisting of mixture of digenite and covellite	22.88
J	XII-D10 Kn 34	Kennecott, Alaska	Medium-grained (~0.5 mm wide) feathered crystals of covellite in a 2–3 mm wide veinlet that cuts a fine-grained covellite host. Veinlet may have formed by hypogene replacement of covellite host although uniform width and uncurved plane of veinlet suggest possibility of small scale fracture filling.	
			Veinlet covellite	21.85
			Host covellite	22.37

DESCRIPTION OF SPECIMENS AND EXPERIMENTAL RESULTS

The analytical results of the isotopic study of the sulfide phases in the specimens mentioned above are listed in the right-hand column of Table 1. The sulfur ratios are also printed on the photographs of some specimens with arrows indicating the various sulfide phases from which these ratios were obtained.

DISCUSSION OF RESULTS

Many suggestions could be made as to why sulfur-bearing minerals in a given deposit might exhibit significant isotopic variations. At this time, however, these suggestions would be based upon numerous assumptions; assumptions on temperatures of formation and what exchange reactions are involved, which in turn are based upon assumptions of the source(s) and composition of the ore-forming fluids, whether equilibrium conditions were attained, extent of isotopic fractionation owing to diffusion and oxidation by meteoric waters, and so forth. The purpose of this initial paper, however, is primarily to indicate that *appreciable variations in* S^{32}/S^{34} *ratios of contiguous sulfide minerals do, in fact, exist.*

Previous Results

Ratios reported by Kulp et al. (10) have shown no evidence of appreciable fractionation of sulfide specimens in juxtaposition. In fact, these investigators have concluded upon initial evidence obtained on reconnaissance data that "the sulfur isotopic abundances within a single deposit appear to be independent of the mineral species and crystal habit at least to $\pm$ 0.2%" (10, p. 148).

Thode and co-workers (19) also have obtained little evidence of appreciable fractionation of closely related specimens although they have reported, for two different specimens, slight variations in the isotopic abundance between fine and coarse crystals from the same pyrite specimens. Fluid-inclusion geothermometry indicates, in the two different specimens described, one from Gilman, Colo., the other from the Larga mine, Zalanthna, Hungary, that the coarse crystals were formed at lower temperatures than the fine crystals. In both specimens, however, the variations are within Kulp's limit of $\pm$ 0.2%. In a footnote, Thode mentions that chalcopyrite crystals from Gilman, Colo., have an S^{32}/S^{34} ratio of 22.261, which differs from the fine-grained pyrite by just slightly more than 0.4 percent. A third pyrite specimen from Sayerville, N. J., described by Thode as "a 'concretion' or round ball of fine crystals embedded in coarse crystals with a thin layer of sulfate mineral between them," exhibits a variation of 3 percent in the concentration of S^{34} between the fine and the coarse crystals. In a recent paper Bateman and Jensen (2) have given a cursory description of two specimens from the Roan Antelope mine, Northern Rhodesia, which although located several hundred feet apart, differ in S^{32}/S^{34} ratio by about 1 percent. Even greater variations are listed in this paper of sulfides from a single hand specimen.

Results of This Study

Simultaneous Paragenesis-Exsolution.—Exsolution or unmixing of specific sulfide minerals takes place, it is believed, by the diffusion to lower energy sites of the small metal cations through the interstices and channelways existing in the structural framework formed by the large sulfur atoms. There is, therefore, a common contemporaneous origin for the sulfur anions forming the framework of closely related sulfides that have undergone exsolution, and those spatially localized sulfide minerals so formed should, therefore, be ex-

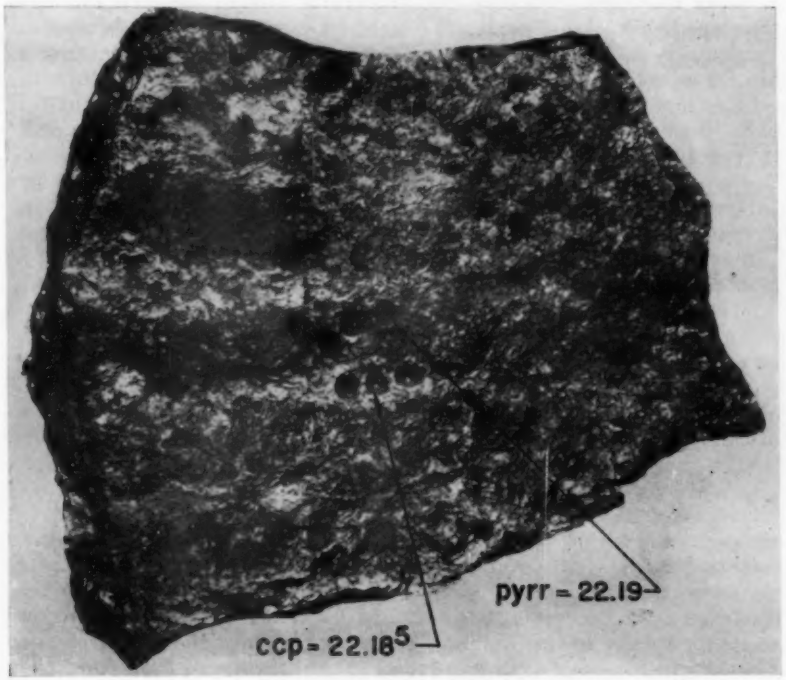

FIG. 1. Alternating coarse bands of chalcopyrite and pyrrhotite. Frood mine, Sudbury. × 1½.

pected to exhibit similar sulfur isotopic compositions. This hypothesis receives support based upon the evidence of similar S^{32}/S^{34} ratios for the two phases analyzed in specimen A from the Mackinaw mine. This specimen was selected, partly because of the ease by which the cubanite and chalcopyrite phases could be separated, but primarily because the two phases exhibit excellent criteria, described in Table I, that indicate an exsolution history.

Penecontemporaneous Paragenesis.—Specimen B, from Sudbury (Fig. 1), contains alternating rather wide bands of pyrrhotite and chalcopyrite.

These bands, some of which are a centimeter wide, are rather large for exsolution lamellae, and therefore may to some suggest magmatic segregation even though the orientation of the bands in the deposit is not horizontal but steeply inclined. In consideration of those who may favor a magmatic origin for this deposit and in light of the relatively large size of the ore body from which this specimen was collected, one might imagine exsolution processes occurring during the slow cooling of this massive sulfide body. However, even if the genesis of this specimen were known, one must recall that Macnamara et al. (13) have shown that there is apparently a rather uniform S^{32}/S^{34} ratio for Sudbury specimens, indicating that appreciable isotopic variation in any of these specimens, even though not formed by exsolution, would be unusual. Incidentally, this rather uniform ratio for Sudbury specimens is becoming broader as isotopic studies continue; it now ranges from 22.08 to 22.19.

Exsolution of chalcopyrite and bornite above a temperature of about 475° C has been reported by Schwartz (17), but the mineralographic textures of the two sulfides in specimen C, from the very low-temperature hydrothermal deposit of Bristol, Conn., are not diagnostic enough to support an exsolution paragenesis. In fact, the limited evidence available suggests either contemporaneous origin or that the chalcopyrite replaces the much more abundant bornite. The S^{32}/S^{34} ratios of the two sulfides, nevertheless, are similar. It is specimen D, described below, which imparts more interest to this specimen.

The large chalcocite crystals from the Bristol deposit are well known. A sulfur isotopic analysis of a portion of one of these crystals, specimen D, has an unusually high ratio of 22.9, in comparison to the ratios of 22.27 and 22.28 of the veinlet specimen C described above. This is a percentage variation of about 2.8 percent for this deposit. Whitehead (23) proposed a supergene origin for these orthorhombic (low-temperature form) crystals. Isotopic studies of sulfur might provide a criterion for distinguishing between supergene and hypogene sulfides. The suggestion, however, that these chalcocite crystals might contain sulfur which was transported in the oxidation zone as sulfate solutions then reduced to sulfide in the supergene zone is puzzling from the isotopic viewpoint to say the least, puzzling because the known partition function ratios of isotopic compounds of sulfur at 0°C and 25°C have been computed by Tudge and Thode (22) and the values of the equilibrium constants for specific isotope exchange reactions, computed from these partition functions, indicate that supergene sulfate solutions will tend to be enriched in the heavier isotope. If the primary sulfide within the supergene zone is entirely replaced by supergene sulfides, the secondary sulfide should exhibit a lower S^{32}/S^{34} ratio. On the other hand, if the replacement process is a diffsion mechanism whereby the metal cations are replaced and the sulfur anions are not replaced, there will, of course, be no change in the isotopic composition. (Actually, there must be a change in the amount of sulfur in some pseudomorphs because of chemical and volume for volume replacement requirements; e.g., pyrite, FeS_2 replaced by chalcocite, Cu_2S.)

The enigma of specimen D (Bristol orthorhombic crystals), of suggested supergene origin, is that it has an S^{32}/S^{34} ratio which is neither lower (enriched in S^{34}) nor essentially the same as the primary veinlet specimen C, but,

instead, is actually very significantly enriched in the lighter isotope. There is, therefore, no isotopic evidence which supports a supergene origin for these crystals.

On the other hand, Bateman (1) has cited strong support for a hydrothermal origin for the chalcocite crystals based upon the knowledge that supergene copper sulfides form by replacement of primary sulfides, and do not form by precipitation or free growth in the supergene zone. Of course, further mineralographic and isotopic studies are continuing on specimens collected from the Bristol mine and upon both supergene and hypogene sulfides from other deposits.

Sequential Deposition.—Needless to say, isotopic similarity of closely associated sulfides does exist in sulfide specimens that lack evidence of an exsolu-

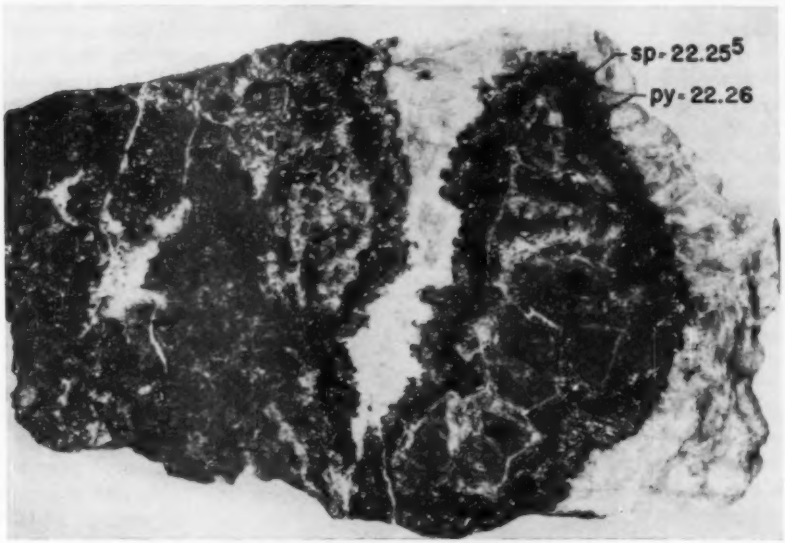

FIG. 2. Cockade ore specimen. Ikuno mine, Japan. × ⅔.

tion history, but instead exhibit evidence of sequential paragenesis. Specimen E, from the Ikuno mine, Japan (Fig. 2), represents sequential or successive deposition of sphalerite, pyrite, and traces of sphalerite as crusts about brecciated gangue and ore specimens, viz., cockade structure. The S^{32}/S^{34} ratios of the sphalerite and pyrite are similar. This may suggest a somewhat rhythmic or sequential precipitation of sphalerite and pyrite from the *same* mineralizing fluid with either slight but significant changes in cation concentrations or possibly sensitive precipitation control under the influence of, for example, temperature and/or pressure fluctuations, instead of deposition from different fluids.

Specimen F, from Galena, Illinois, (Fig. 3) may have had a history similar to specimen E described above in that an early sulfide has been covered with a crustification of a second later sulfide. Both minerals are primary, and the perfect cubes of galena showing no evidence of oxidation or leaching does weaken the possibility of supergene processes forming the marcasite crust

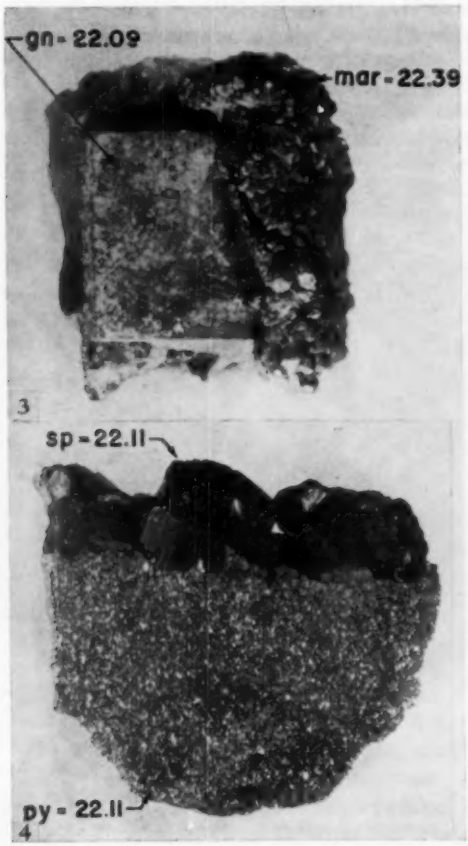

FIG. 3. Galena coated with marcasite. Marsden's name, Galena, Ill. × 3.
FIG. 4. Sphalerite crystals on pyrite pedestal. Vigsnus, Norway. × 1.

covering the galena. In contrast, however, there is a S^{32}/S^{34} ratio variation of almost 1.4 percent between the galena (22.09) and the marcasite (22.39^5). Whether an appreciable temperature rise, isotopic fractionation processes, different sources of mineralizing fluids, or combinations of these factors brought about the isotopic variation is not known. Further studies, both in the

laboratories in synthesizing sulfide minerals under controlled conditions, and in the field where a detailed geologic study may indicate which processes are more probable, are needed in deciding the relative merits of these various factors.

Specimen G, from Norway, (Fig. 4) also exhibits two sequential periods of formation of different sulfides, an early stage of pyrite followed by the later growth of coarse crystals of sphalerite. Although there are periods of successive deposition of sulfides in both specimens F and G, the latter in contrast shows no S^{32}/S^{34} variations between the pyrite and sphalerite, both with ratios of 22.11.

A relatively large vein specimen, specimen H, (Fig. 5) consisting of sulfides and quartz gangue was the first sample selected in which significant isotopic variations were noted. Galena was formed first, followed by deposition of milky-white quartz, which was followed by deposition of sphalerite, then galena, and finally a mixture of several fine-grained sulfides and jasper. The later galena and sphalerite exhibit S^{32}/S^{34} ratios of 22.19 and 22.21 respectively. Significantly, however, the early galena stage that is separated from the later sulfide history by a quartz stage, has a ratio of 22.39. Once again, the question of the cause of the variations arises. The relatively slight isotopic fluctuation in the later sulfides, although just within the allowable precision of results, might be due to temperature fluctuations. These two ratios, which may truly differ only slightly, might be expected from *theoretical* considerations of the *theoretical* equilibrium reactions that do show a lesser fractionation effect at higher temperatures per degree than near the range of room temperature. (There is, obviously, a need for experimental verification of these theoretical beliefs.) A somewhat different reason, such as the source of mineralizing fluid, may best explain the different ratio of the early galena, especially since it is separated from the later sulfides by an interrupting quartz stage.

Replacement Mechanism.—Specimens I and J were selected from the well-studied (3, 4, 11) Kennecott, Alaska, suite. Specimen I (Fig. 6), is noteworthy because it shows the largest variation in S^{32}/S^{34} ratios thus far measured in one specimen. The coarse-grained veinlet of covellite with a ratio of 21.91 encloses a few small veinlets composed of a mixture of mostly digenite and some very fine-grained covellite, which has a ratio of 22.88. This is a variation of more than 4 percent. Furthermore, one of the digenite walls that encloses the one-inch wide covellite veinlet has a ratio of 22.64. The specimen is apparently entirely of hypogene origin with no evidence of supergene processes. Specimen J, also from Kennecott, consists essentially of relatively fine-grained covellite, with a S^{32}/S^{34} ratio of 22.31, cut by a later coarse-grained veinlet of covellite that has a ratio of 21.85.

This coarse-grained covellite veinlet in specimen J is uniformly thin and tabular in shape and, therefore, may not have formed by replacement but by simple filling of a planar fracture. In specimen I, however, the mutual boundaries and rapid fingering-out of veinlets indicate that they have formed not by fracture filling but by replacement of the existing sulfide host.

Lindgren (12) almost half a century ago, wrote that "the difficulty of an

FIG. 5. Portion of crustified vein; wall rock at left, next galena, then quartz, sphalerite, galena, and fine-grained sulfides in jasper. × ½.

FIG. 6. Digenite host borders polished surface of coarse-grained covellite vein cut by replacement veinlets of chalcocite and a mixture of fine-grained chalcocite and covellite. × 2.

adequate explanation of the geologic process known as metasomatism or replacement has long been recognized." The space problem in particular was difficult to explain adequately. One of Lindgren's students, M. J. Buerger (5), offered a very reasonable partial explanation as follows: "Lindgren long ago pointed out that replacement occurs on approximately a volume-by-volume basis. While the field evidence for this has been obvious, the mechanism for accomplishing it has been obscure. Diffusion suggests the mechanism. There is a tendency on the part of crystals to have their volumes determined by their largest atoms. Thus, the volumes of the rock minerals are dominated by their oxygen atoms and the volumes of the sulfides are dominated by the packing of the sulfur atoms. Replacement is, therefore, substantially a matter of the diffusion of new metals into the volumes dominated by oxygen or sulfur atoms. Thus, diffusion supplies a mechanism for approximately maintaining volume during replacement." In this explanation of the process of replacement there is no exchange of the sulfur atoms, but in both specimens, I and J, there is no similarity in the isotopic composition of the original host sulfide and the replacing sulfide. Quite evidently, therefore, in these specimens, possibly with the exception of specimen J if it represents fissure filling, there has not only been an exchange (in concentration) of the metal cations but also an exchange of the large sulfur anions. At the present time, we are continuing further studies on this interesting and important finding.

Incidentally, it does seem that instead of a diffusion mechanism, a mechanism of dissolving of host followed by precipitation of replacing mineral described by Lindgren (12) would more easily explain why the sulfur atoms are also replaced; i.e., "that complex reactions constantly take place in the contact films of solutions; the process goes on in one operation, solution and precipitation following so closely that no spaces of solution are visible under the microscope." Nevertheless, and parenthetically, there is little doubt that solid state diffusion is still an important process in exsolution, replacement, and the formation of intermediate phases in the realm of sulfide mineralogy, as indicated by evidence provided by Ross (16), Frueh (6, 7), and Jensen (8).

CONCLUSIONS

In the previous section, the specimens have been divided into four groups in which the two or more sulfides within each specimen vary in paragenesis from simultaneous, through sequential, to non-contemporaneous respectively. The *a priori* assumption, borne out by the results, that there is a greater probability of variations in S^{32}/S^{34} ratios in the latter groups is not surprising because the assumption is based essentially upon the significance of time dependency; i.e., the greater the difference in time of origin of adjacent minerals, the more probable it is that they will differ in isotopic composition.

It is clear that significant and appreciable S^{32}/S^{34} variations do exist in some contiguous or closely related sulfide minerals, sulfides not only of different species but of even identical species. Adequate explanations of the processes that have led to the formation of hypogene sulfides of differing isotopic composition are lacking, although slight variations, of the order of a few hundredths, may be dependent on temperature changes. Similar and even

greater variations may be non-equilibrium fractionation or unidirectional dependent processes. Variations of the order of a few to several tenths suggest a more complicated "plumbing system" for the passage of hydrothermal solutions and possibly more than one distinct source for the solutions; for example, the ore-bearing solutions may be tapped from several localized and coalescing ore-fluid concentration zones. A cyclic hydrothermal plumbing system giving a cumulative fractionation might provide the panacea needed by the theorist to explain the measured extensive fractionation of the sulfur isotopes, but this is obviously an *ad hoc* hypothesis.

Even the most casual reader is now most certainly aware that mere theorizing based upon isotopic studies of hand specimens picked from mineralogic collections is no way to continue this study. Instead, what will be more enlightening is to carry out a detailed geologic examination (à la Dr. Livingstone!) of a not too complex but accessible hydrothermal mineral deposit in which primary sulfides have and are still undergoing oxidation. Accessibility to both the enriched supergene zone, in which the primary sulfides have not been entirely replaced, and the underlying protore should provide adequate means for obtaining further knowledge about the possibility and extent of natural oxidation processes and the resulting extent of fractionation of the sulfur isotopes. The study would be further facilitated by collecting sulfate-bearing mine waters at various zones within the deposit. Initial arrangements and skeletal collections as briefly outlined above have been obtained and are to be continued and greatly expanded by this laboratory.

Furthermore, the various processes of exsolution, solid solution, replacement, the formation of intermediate sulfide phases, and even an understanding of what some investigators believe to be the comparatively simple process of growth of sulfide minerals, should be further studied in view of the additional knowledge to be gained by use of the mass spectrometer. The formation of synthetic sulfide minerals by various processes under controlled but different conditions of temperature and pressure from sulfur of known isotopic composition might provide information of substantial interest.

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I am greatly indebted to Professor Alan M. Bateman not only for his invaluable aid and assistance in discussion of the various problems encountered and some of the thoughts presented in this study but also for his foresight and faith in expanding the Laboratory of Economic Geology to include equipment and facilities needed for the application of the subject of isotope geology to the realm of economic geology.

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SHEARING ALONG ANTICLINES AS AN IMPORTANT STRUCTURAL FEATURE IN URANIUM MINERALIZATION IN THE NORTHERN PART OF THE NORTHERN TERRITORY OF AUSTRALIA

J. RADE

ABSTRACT

Deep seated flow of geoplasma (bathyrheal underflow), has been responsible for horizontal movements of crustal blocks of Lower Proterozoic in the northern part of the Northern Territory, Australia. These movements resulted in strike-shearing of anticlines in the Lower Proterozoic Brock's Creek Group, and the resultant shattered and sheared anticlines became the locale for uranium mineralization. These strike-sheared anticlines are an important guide in uranium prospecting in the northern part of the Northern Territory.

INTRODUCTION

SHEARING along anticlines is an important structural feature associated with uranium mineralization in the northern part of the Northern Territory. The structure is well seen in the area between 131°E and 132°E and 13°S and 14°S which is within the Darwin-Katherine region of Noakes (6) and the Rum Jungle Province of Fisher and Sullivan (1).

The area is mainly occupied by siltstones, sandstones, conglomerates and tuffs of the Lower Proterozoic Brock's Creek Group (6). At the end of the Lower Proterozoic these were folded and intruded by granite, which now outcrops as the Mt. Shoobridge, Brock's Creek and Prince's Springs Granites. The granite bodies represent separate crustal blocks and their horizontal movements have played an important part in the formation of the structural feature described in this paper. The Brock's Creek Granite is the largest of these outcrops and the Mt. Shoobridge Granite is the smallest. In the southeastern part of the area the large massif known as the Cullen Granite (6) outcrops.

The shearing along anticlines that is connected with uranium mineralization occurs in rocks of the Brock's Creek Group. Two typical examples will be described.

EXAMPLES OF SHEARING

Burrundie Prospect.—The first is the Burrundie Uranium prospect 95 miles southeast of Darwin and 3¾ miles southwest of Burrundie railway siding. The sedimentary rocks here are partly hornfelsed grey siltstones, which are in some places bleached to white, belonging to the Brock's Creek Group. The siltstones contain pyrite and cavities one-eighth to one-quarter of an inch in diameter have been formed by solution of the pyrite. Quartz has

been injected as veins and lenses and the larger lenses contain vugs filled with quartz crystals. The prospect is surrounded by granite, the Prince's Springs Granite to the north and the Cullen Granite to the south.

Both regional and local folding and faulting affected the rocks of the Brock's Creek Group in the Precambrian. The Burrundie Uranium prospect is located on the crestal part of an anticline that has been subjected to tension during folding, as have many of the other anticlines in this area. In this case tension fractures were formed and kept open, allowing mineralizing solutions to circulate. Quartz was injected along the tension fractures during the last stages of igneous intrusion. Later the crestal part of the anticline was affected by shearing due to horizontal movements in the basement. The quartz veins were brecciated and shattered and hematitic solutions circulated, filling the interstices between the broken quartz grains with hematite.

Recurring movements have developed intense fracturing in the Burrundie Uranium Prospect. The main trends of the fractures are very characteristic. Both shear and tension fractures are developed, which clearly show that the northeastern part of the anticline moved to the southeast and the southwestern part moved northwest. Some cross-fractures trending northeast transect the crest of the anticline. Their positions are indicated by the topography, the largest quartz veins, associated fracturing and the hydrothermal alteration of the rocks. Such cross-fracturing is a common structural feature. Gilkey (2) has described it in several anticlines in the United States of America; for example, of the anticline in the Marathon Region, Texas, he says that "the most continuous set of joints can be seen to parallel in general the direction of the faults and to intersect the axis of the fold at about seventy-five degrees, in one direction only." Further on he discusses the faulting and jointing in the Wills Mountain Anticline at Maryville Gap, Central Appalachians, West Virginia, and says: "One characteristic of the density of the jointing was apparent in the field, the very marked increase in density toward the crest of the fold. For several hundred feet above the bottom of the gap, the orientation is less pronounced and the jointing is relatively sparse, while above that, the density increases to almost a sheetiness along the highest part of the fold." The cross-fracturing at the Burrundie Uranium prospect is similar to that described by Gilkey. The cross fractures commonly form either during or after the folding of the anticlines. Diorite and amphibolite encountered in the core of the anticline were intruded during folding, and the quartz that fills some of the cross-fractures represents the last product of differentiation of the same magma.

The author believes that some slight movements along the northeasterly trending cross fractures were rejuvenated at a later date. This is shown by shearing in the quartz-filled cross fractures in the southeastern and northwestern parts of the Burrundie Uranium prospect. Fractures show that the northwestern parts of the cross fractures moved to the northeast and the southeastern sides to the southwest. It should be noted that similar weak movements along northeast trending cross fractures are characteristic of the whole of the northern part of the Northern Territory. On a large scale, the same type of movements affected the crustal blocks in the eastern part of the

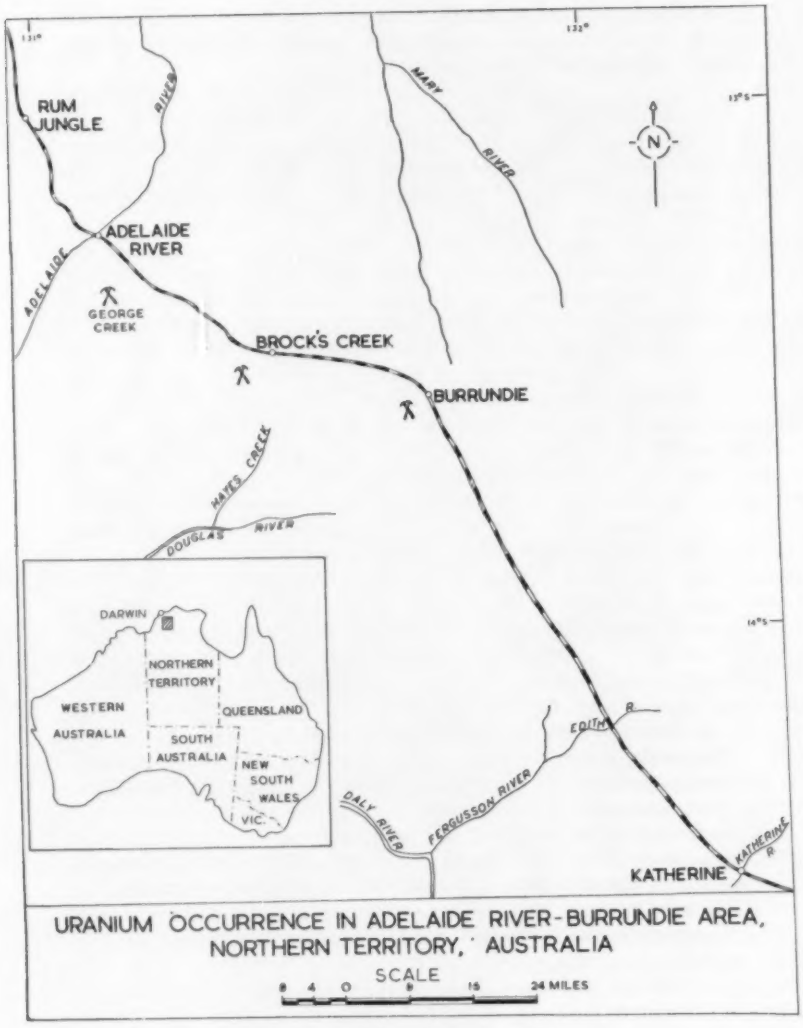

FIG. 1.

Pine Creek geosyncline, which includes the area under consideration. The author believes that the cause of these horizontal movements was a deep-seated flow of geoplasma—the bathyrheal underflow (7).

The radioactive deposits at Burrundie are of hydrothermal origin. Patches of radioactive limonitic gossan were found in several places on top

of the razorback ridge on which the prospect is situated. The gossan material is a dirty brown color, light in weight and highly leached because its position on the razorback ridge is favorable for leaching. Not all the gossans of this type were found to be radioactive. The radioactive gossans tend to occur where the fracturing is strongest at the intersections of the shearing in the anticline with the northeasterly trending cross fractures. Thus the cross-fracturing is an important factor favoring uranium mineralization in this case. It has been observed overseas and also by the author in the Northern Territory that radioactive mineral deposits favor highly shattered zones, especially those in brittle rocks. An overseas example is the Precambrian Red Creek Quartzite in the northern part of the Uinta Mountains, Daggett County, Utah (10), and in Australia the carbonaceous slates and graphitic schists at Rum Jungle, Northern Territory (9) and the schists of the Sleisbeck area, Northern Territory (5) are other examples.

It is characteristic of uranium mineralization that zones of minor fracturing are preferred to major fault zones. An example of this is in the Goldfields Region, Canada (4) where only a few deposits have been found within large fault zones, the principal ones being in zones of minor faults, crushed zones, or in tension fractures near the major faults.

Shear zones are commonly favored in the deposition of radioactive minerals. The mineralization is commonly a late phase, occurring after the minor movements that cause the brecciation and intense fracturing. Such a case is described by Hart and Hetland (3) from the Mohave County, Arizona, where the last movements were weak in comparison with the earlier ones. The Burrundie area is similar in that several movements occurred, as shown by slickensides and brecciation of quartz veins, the later movements being weaker than the earlier ones. The mineralization accompanied the later movements.

The mineralization and the associated movements in the Burrundie area may be summarized as follows:

1) Folding of grey siltstones with the formation of an anticline. Formation of tension fractures in the crest of the anticline; cross faulting in the anticline.

2) Injection of quartz along tension fractures and the larger cross fractures in the crestal part of the anticline.

3) Mainly horizontal movements in the basement causing shearing in the overlying sediments with shear and tension fractures in the crest of the anticline and brecciation of quartz veins.

4) Intrusion of hematitic solutions into the brecciated quartz veins and tension fractures.

5) Slight shearing along the cross-fractures.

6) Radioactive mineralization of the crestal part of the anticline, the mineralization favoring the regions of strongest fracturing and the intersections of the northeasterly trending cross fractures with the crest of the anticline.

7) Hydrothermal alteration of some parts of the crest of the anticline contemporaneously with the mineralization (6).

It should be noted that the brecciated quartz veins are not radioactive; only the highly fractured gossan is radioactive. From this it is seen that the porosity or degree of fracturing of the vein material has a definite influence on the distribution of radioactive minerals in the Burrundie Uranium prospect. Hart and Hetland (3) have made similar observations in Mohave County, Arizona; they say: "When uranium occurs in a vein a definite porosity is noted, and where a decrease or absence of radioactivity is apparent, the vein matter is tight or not shattered." Thus the shattering of the rocks is seen to be an important factor in uranium mineralization. In the Burrundie area the anticlines were fractured during folding, but subsequent shattering by shearing movements was important in the mineralization of the anticline.

Since the shearing of anticlines in rocks belonging to the Lower Proterozoic Brock's Creek Group is so closely connected with uranium mineralization in the Northern Territory the presence of this structural feature was suggested by the author as a reliable guide in prospecting for uranium. In 1954 this led to the discovery by the author of the George Creek Uranium Prospect, which is situated 63 miles south-south-east of Darwin and 8 miles south of Adelaide River (7). The country rock consists of greywackes and siltstones of the Brock's Creek Group, showing cyclic sedimentation and common slump structures. Coarse greywackes grade into fine greywackes and siltstones, and banded light and dark grey siltstones are intercalated with them.

George Creek Prospect.—The George Creek Uranium Prospect is situated on the side of an anticline trending approximately north-south, which has been sheared along its length, the east side moving north and the west side south. Characteristic tension and shear fractures are present. Uranium mineralization has favored tension fractures in the strongly shattered siltstones. Pitchblende has been encountered in the tension fractures and it is from these fractures that uranium has been introduced into the banded siltstones with partial replacement of beds. Here again in the George Creek Uranium Prospect, strong shattering caused by folding and strike-shearing has favored uranium mineralization. The main trends of this fracturing are shown on the surface geological map of the prospect (Fig. 2).

Another uranium prospect that belongs to the same type is the Brock's Creek Uranium Prospect located 80 miles SE of Darwin and 26 miles SE of Adelaide River in the "gold-zone" of Sullivan and Iten (8). It is located on a strike—sheared anticline intruded by amphibolite. The north-eastern side of the anticline has moved to the NW and the south-western side to the SE, the movement being accompanied by strong fracturing and shattering. The rocks exposed at this prospect belong to the Lower Proterozoic Brock's Creek Group.

Conclusion.—In the present paper the author has attempted to interpret the association of uranium mineralization with sheared anticlines in the northern part of the northern Territory. He believes that movements of crustal blocks in this area have been caused by bathyrheal underflow and that these movements favored uranium mineralization in the resultant structural

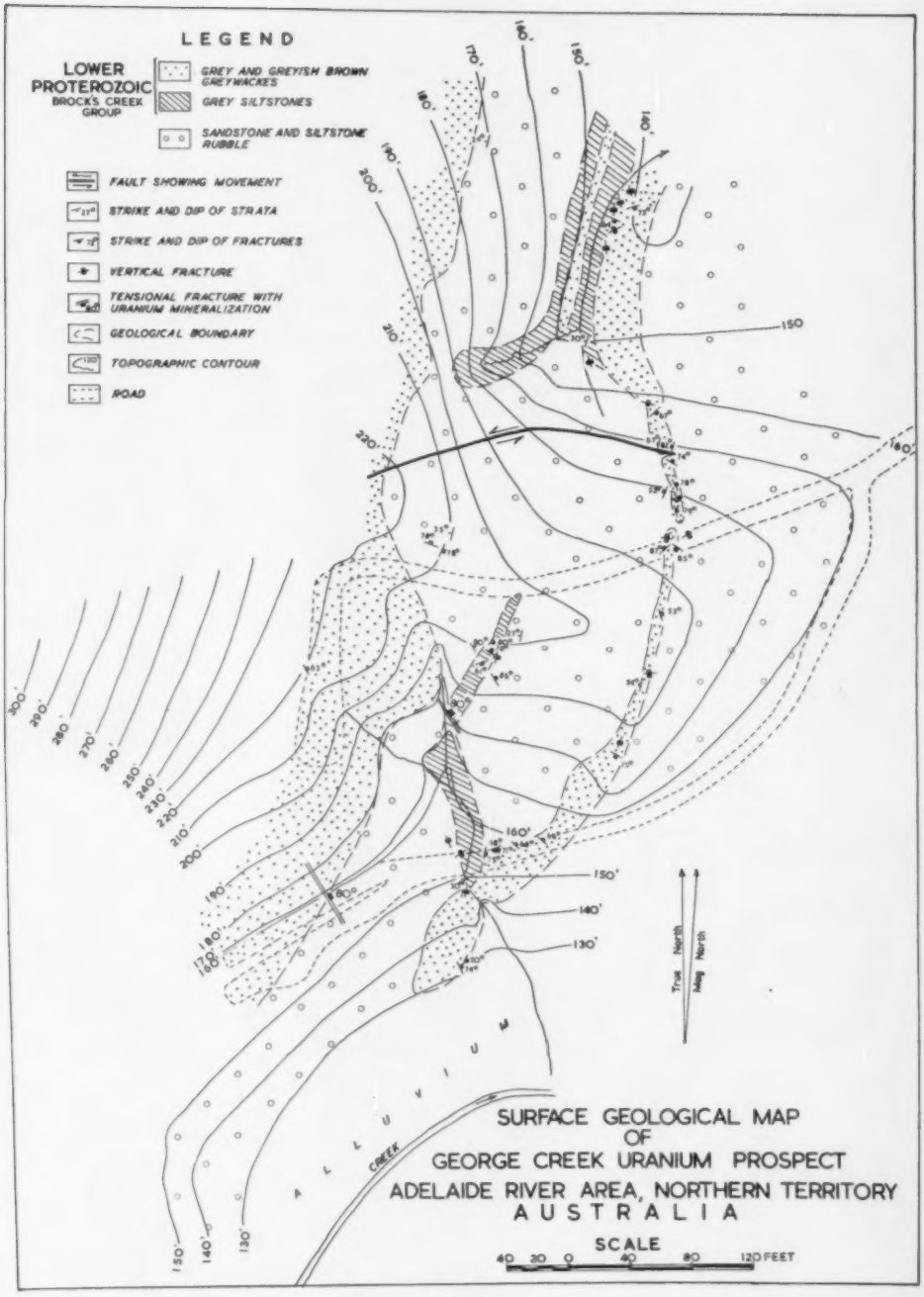
LEGEND
LOWER PROTEROZOIC
BROCK'S CREEK GROUP
GREY AND GREYISH BROWN GREYWACKES
GREY SILTSTONES
SANDSTONE AND SILTSTONE RUBBLE
FAULT SHOWING MOVEMENT
STRIKE AND DIP OF STRATA
STRIKE AND DIP OF FRACTURES
VERTICAL FRACTURE
TENSIONAL FRACTURE WITH URANIUM MINERALIZATION
GEOLOGICAL BOUNDARY
TOPOGRAPHIC CONTOUR
ROAD
ALLUVIUM
CREEK
True North
Mag North
SURFACE GEOLOGICAL MAP
OF
GEORGE CREEK URANIUM PROSPECT
ADELAIDE RIVER AREA, NORTHERN TERRITORY
AUSTRALIA
SCALE
40 20 0 40 80 120 FEET

FIG. 2.

features. Consequently, future search for uranium should be concentrated on such structural features as sheared anticlines.

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Aug. 15, 1956

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A TRUCK-MOUNTED SPECTROGRAPHIC LABORATORY FOR USE IN GEOCHEMICAL EXPLORATION [1]

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ABSTRACT

A truck-mounted spectrographic laboratory has been designed and built by the U. S. Geological Survey to investigate the feasibility of using and transporting such equipment in the field as an aid in supplying rapid on-the-spot analytical data to geochemical exploration field parties.

The laboratory is housed inside a 7- × 12-foot insulated and air-conditioned van-type truck body and carries complete equipment for making qualitative, semiquantitative, and quantitative analyses of geological materials. The spectrograph is a fixed-position 1.5-meter grating instrument of the Wadsworth type which records a range of spectra from 2063 to 4837A in the second order on a 20-inch strip of film. Companion units are a projection type comparator-densitometer, a film processor, and other accessory equipment. Trailer-mounted motor-generators supply 230 volts d-c for the arc source unit and 115 volts a-c for lights and accessories.

Since its completion in May 1955, the truck-mounted laboratory has been driven over 4,000 miles. During this travel all the equipment remained in good adjustment; the laboratory was made ready for operation in less than 2 hours after arrival at the site of a field project.

Because of the large number of elements that can be determined from a single spectrogram, the truck-mounted laboratory is useful in the early

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stages of a geochemical exploration project to determine diagnostic suites of elements, and later, to guide the day-to-day sampling.

INTRODUCTION

GEOCHEMICAL prospecting techniques are in common use today in the search for concealed mineral deposits. These techniques usually require that a large number of samples be analyzed quickly, and preferably during the geochemical survey so that any geochemical anomaly present may be known and evaluated at once. Rapid and inexpensive analytical methods for determining traces of metals are essential, and portability is a desirable feature of such methods because geochemical exploration programs are often hampered by the lack of rapid, on-the-spot analytical data, especially in the earlier phases of such programs. Portable analytical equipment and procedures that can be used near the site of the sampling program allow the field men to receive data on the metal content of their samples promptly and permit more efficient planning of the day-to-day collection of samples. To date, in the United States, most of the portable analytical methods are chemical procedures that were developed especially for use in geochemical prospecting (3). These methods have been largely responsible for geochemical prospecting attaining the status of a recognized and successful exploration tool.

Geochemical prospecting is relatively new, and as is so often true of a new technique, it is still largely empirical in nature. Thus, before embarking on any major geochemical survey, preliminary orientation studies are almost mandatory in order that diagnostic suites of elements be determined and other geochemical and sampling problems evaluated. It is not necessarily true, for example, that the sought-for metal in a deposit is the one that will be the most valuable in deciphering the geochemical dispersion pattern.

Spectrographic methods of analysis are most useful in the orientation phases of a geochemical program when the diagnostic elements are not yet known with any degree of certainty, because many elements can be determined from a single sample with little more effort than that required to determine one. They should also be the most enconomical in those surveys where each sample must be analyzed for a considerable number of elements. The chemical methods developed for use in geochemical prospecting are less generally useful and more tedious in this phase of the project.

Spectrographic methods have played a minor role compared to the wet colorimetric methods of trace analysis in geochemical surveys in this country, but in the Union of Soviet Socialist Republics and the Scandinavian countries spectrographic procedures were used extensively in the early geochemical prospecting work carried on prior to World War II.

Scientists of the Union of Soviet Socialist Republics also pioneered in the development of mobile spectrographic laboratories for accompanying and servicing geochemical prospecting parties. In 1939 Ratsbaum (4) mentioned that two types of spectrographic laboratories had been used in the field by Soviet geochemists since 1935, and stated that the organization of a mobile laboratory mounted on a truck was entirely feasible. He listed the equipment required but gave no details about actual organization and arrangement of the equip-

ment in the truck. Although it is to be expected that Soviet geochemists have continued the development of mobile spectrographic laboratories, no recent information is available about their activities in this field.

In 1952, H. E. Hawkes, then Chief of the Geochemical Prospecting Section of the U. S. Geological Survey, proposed that the Survey investigate the applicability of spectrographic procedures to the field of geochemical prospecting. To facilitate this study, plans were made to construct a mobile laboratory that could be driven to the vicinity of field operations and used there for servicing geochemical prospecting field parties. This laboratory was completed in May 1955 and first tried out in the field in June 1955. The results of experience with this unit between June and December 1955 were considered favorable enough to warrant publication of a report covering a description of the equipment and its performance in the field. The spectrographic methods used will be described in a separate publication.

As no discussions of theory and general principles will be included in this paper, readers who are interested in the basic theory and the applications of spectroscopy are referred to Harrison, Lord, and Loofbourow (2), and Ahrens (1). The reference to Harrison et al., covers the entire field of spectroscopy and discusses both basic theory and instrumentation. Ahrens, on the other hand, deals more specifically with the problems that arise in analyzing geological materials.

ACKNOWLEDGMENTS

In the design and construction of this unit we benefited from the constructive advice of many of our colleagues. Many thanks are also due to the staff of the Geological Survey machine shop at the Denver Federal Center, especially C. G. Bay and W. H. Wood, for their wholehearted cooperation and assistance.

DESIGN CONSIDERATIONS

The over-all aim in designing this laboratory was to place all of the equipment required to make qualitative, semiquantitative, and quantitative spectrographic analyses of geological materials into a truck that could be driven to or near the base camp of the geochemical prospecting field project so that rapid on-the-spot analytical services would be immediately available. Such a mobile laboratory would be most useful if it were completely self-contained and capable of operating for appreciable periods of time without benefit of such urban facilities as power and water supplies. This aim was achieved and the present laboratory can be operated in remote areas for long periods, provided supplies of gasoline can be received periodically.

The design problem consisted of two separate phases: first, suitable items of equipment had to be obtained; and, second, these items had to be arranged and mounted in the truck so that they would be protected from shock and vibration damage while the truck was moving.

The basic criterion in selecting equipment for this unit was to produce a spectrographic laboratory at a minimum cost consistent with obtaining the analytical precision considered necessary for most geochemical prospecting

surveys. No attempt was made to design a laboratory that could compete in precision and accuracy with the elaborate spectrographic equipment found in many stationary laboratories. Rather, we envisioned a unit that would be the spectrographic analog of the chemical field methods developed by the Geological Survey to meet the practical requirements of geochemical prospecting. Similarly, the methods used in this mobile laboratory were engineered and adapted for use under field conditions by sacrificing some precision and accuracy in favor of speed, simplicity, and economy.

It was also obviously desirable that the spectrographic equipment and other components should be standard items readily available from commercial manufacturers. Because of space limitations, the necessity for conserving

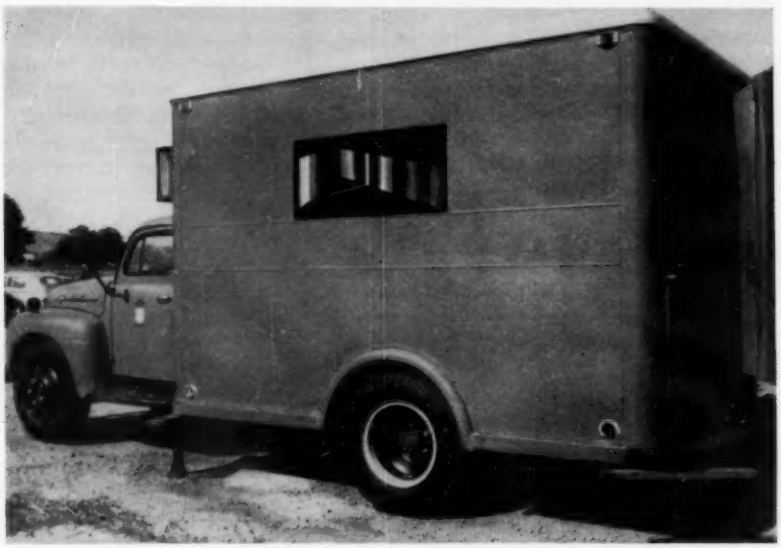

FIG. 1. Exterior view of laboratory truck.

water, and other factors, this desideratum was not always achieved, but the source unit for the arc was the only major item constructed in its entirety. A number of major items of equipment, such as the spectrograph and film processor, that were modified by the manufacturer to meet our requirements have since been placed on the market in their modified form.

DESCRIPTION OF LABORATORY

The laboratory is housed inside a standard but specially insulated 12-foot van-type truck-body mounted on a standard 2-ton truck chassis. Electrical power for the laboratory is supplied by two motor-generator plants that are mounted on a small 2-wheel trailer which is towed by the truck. Figure 1

shows an exterior view of the laboratory truck, and Figure 2 shows the mobile electrical power plants.

Truck Body.—A standard 12-foot van body was modified by placing two inches of fiberglass insulation along the sides, ends, and ceiling to help maintain a controlled temperature under varying climatic conditions. This insulation is covered by panels of plywood.

A 1/2-horsepower room-type air conditioner is mounted in the upper left forward end of the body. A corner of this unit is visible above the truck cab in Figure 1. This unit was installed to allow the laboratory personnel to work efficiently in hot areas and not for maintaining the spectrographic

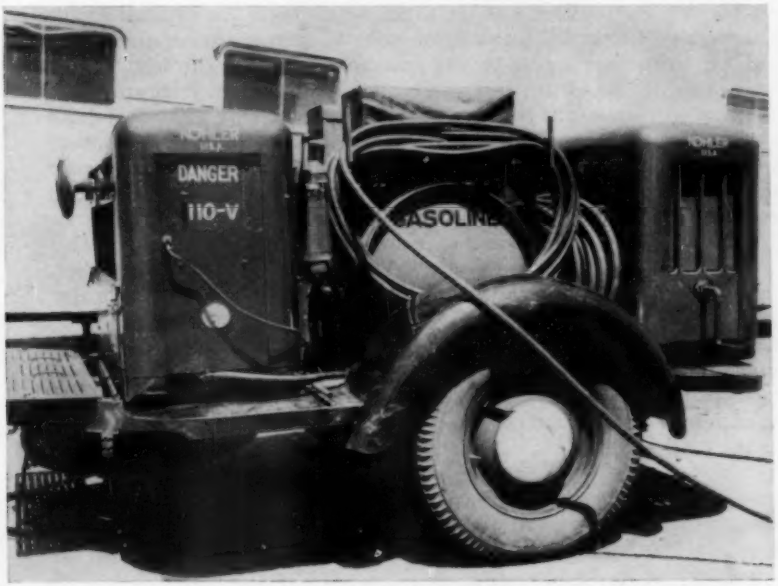

FIG. 2. Mobile electrical power plants.

equipment at a constant temperature. The unit can be set to bring in fresh, filtered outside air, either cooled or uncooled; to exhaust room air to the outside; or provide maximum cooling by recirculating and filtering the inside air. A portable electric heater is used during the winter months to heat the interior of the body.

Mobile Electrical Power Plants.—The electrical plants consist of one 5-kw 230-volt d-c generator, and one 3.5-kw 115-volt 60-cycle a-c generator. Each generator is driven by a 4-cylinder water-cooled manual-start gasoline engine. Both generating plants are equipped with automatic voltage regulators that limit voltage fluctuations under normal load conditions to within about 5 percent of the rated output voltage. Both plants are also equipped

with weatherproof metal housings, oil filters, and air cleaners, which permit operations under adverse weather conditions. Running-time meters on each plant record the time operated. Each unit is provided with 100 feet of cable to connect it with the appropriate circuit within the truck. Different types of polarized plugs are used to prevent the interconnection of the 230-volt d-c generator and the 115-volt a-c truck circuit. The 230-volt d-c plant is used mainly for supplying power to the arc source unit, although a number of minor accessory items such as the laboratory oven and mortar grinder operate on this voltage. The 115-volt a-c plant supplies power for the lights and the majority of the accessories.

The two electrical plants are mounted on a 2-wheel steel-bed trailer of 1-ton capacity. A 55-gallon gasoline drum, which holds the gasoline supply for both plants, is mounted between the units in such a manner that it can be removed to provide access to the generating plants. The load on the trailer is so well balanced that two men can easily move it on level ground.

Electrical Circuits.—The laboratory is equipped with both 230-volt d-c and 115-volt a-c electrical circuits, which are placed inside the thin-wall electrical conduits mounted on the plywood panels. Outlets are located at appropriate intervals and different types of plugs and sockets are used on each circuit to prevent connection of 115-volt equipment to the 230-volt line, and vice versa. Both circuits contain a third wire that is used as a ground connection. This wire is connected to a heavy copper ground rod carried on the power trailer. This rod is driven into the earth when the laboratory is operating. All major accessories have been equipped also with a ground wire so that connecting any such accessory to an electrical circuit grounds it automatically.

An instrument panel containing voltmeters and ammeters for both circuits is located on the rear wall just above the work bench to permit convenient checks of the voltage and current in any circuit. In an emergency, all power can be cut off from both circuits by main switches located on the rear wall panel.

Arrangement of Equipment.—The general arrangement of the laboratory is shown in Figures 3, 4, and 5. A work bench, 21 inches wide, and 31 inches from the floor, extends along the entire left side of the truck interior. Two working sites are provided at this bench, one at the extreme forward end, extending underneath the air conditioner, and the other at the rear by the door. Electrodes are loaded, and samples and standards are prepared in the forward location out of the way of traffic between the arcing bench of the spectrograph and the darkroom door. Directional fins on the air conditioner can be adjusted to deflect the flow of air away from this area to avoid interference with weighing operations. The rear working location is used for examination of films, preparation of reports, and other miscellaneous paper work. A small electric oven and a motor-driven mortar grinder are mounted on the bench above the water tank. A storage compartment is mounted on the side wall above the oven and mortar grinder.

A stainless steel 70-gallon water tank, $38 \times 21 \times 20$ inches, rests on the left wheel-well under the center part of the bench. This tank supplies water to the darkroom for film washing. A 50-gallon auxiliary water tank, $22 \times$

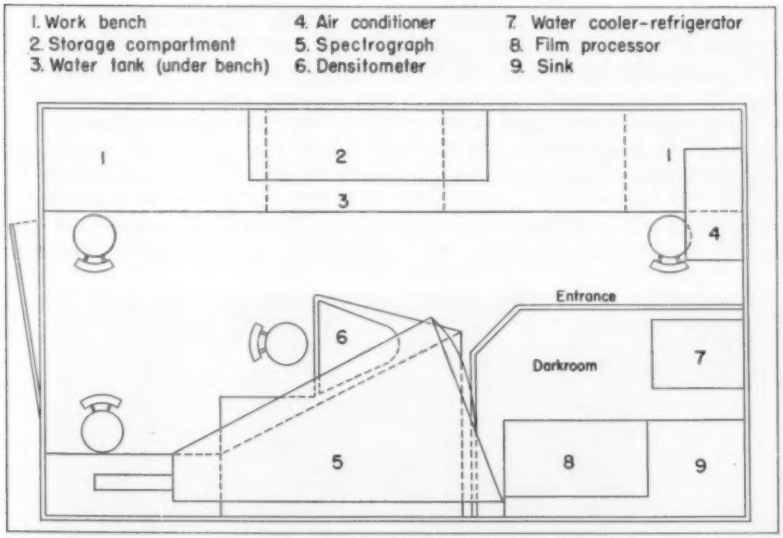

FIG. 3. Location of major items of laboratory equipment.

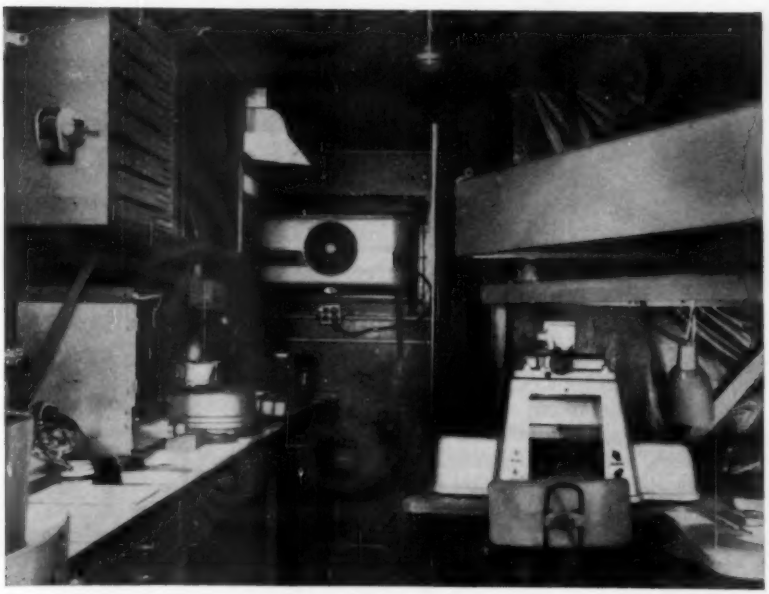

FIG. 4. General interior view.

48 × 11 inches, is mounted beneath the floor in the left forward corner. Baffle plates in both tanks minimize surging of the water during travel. The total 120-gallon capacity of these two tanks permits the laboratory to operate for extended periods before additional water supplies are required. Both tanks full of water serve to balance the weight of the spectrograph and other heavy items, which are mounted on the right side of the truck. The weight distribution becomes important when the unit is moving over high-crowned roads.

On the right side of the truck, the spectrograph is mounted 50 inches above the floor on a wedge-shaped bench. One corner of the spectrograph projects

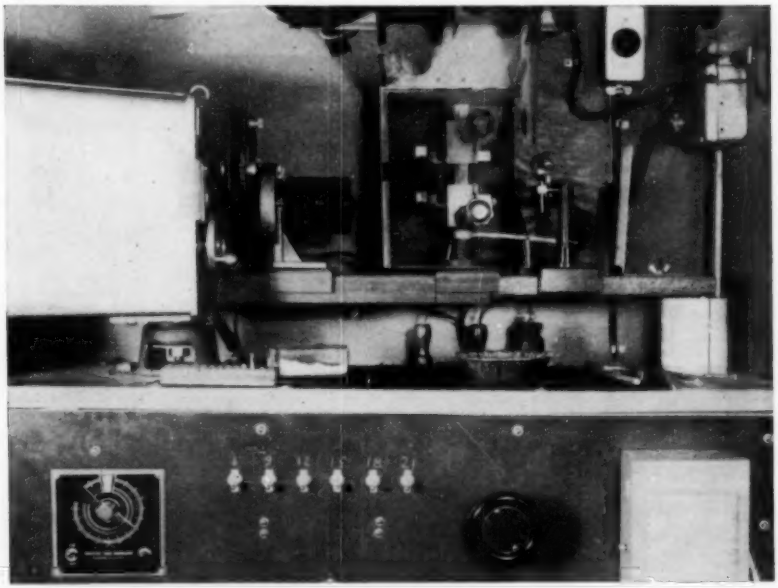

FIG. 5. Optical bench and control panel.

into the darkroom through a light-tight opening in the darkroom curtains. The optical bench and control panel for the arc source unit are located in the right rear corner of the truck. Much of the wiring of the source unit is placed behind the control panel beneath the arcing bench; however, the major banks of heat-producing resistors are mounted in a separate compartment located beneath the truck floor and opening to the outside. The arc fumes are exhausted through an opening in the wall just below the truck roof by a small fan in a canopy hood fastened above the arc stand. The comparator-densitometer is on a lower table beneath the spectrograph.

The darkroom is in the right forward corner of the truck and is partitioned from the remainder of the interior by light-tight curtains. Inside the dark-

room, a bench along the right side holds a film processor on one end and a small cup sink and faucet at the other end. A combination water cooler-refrigerator provides cold water for the film processor and refrigerated storage for film.

Spectrographic Equipment.—In 1952 when the initial plans were made to construct a mobile spectrographic laboratory, procurement of a suitable spectrograph was the first problem that had to be faced. Although prism spectrographs are sometimes considered to be more rugged and portable than grating instruments, a consideration of the relative merits of grating and prism spectrographs left no doubt but that a grating spectrograph would be the most suitable instrument for the mobile laboratory. Harrison et al. (2, p. 27–50) give an excellent discussion of the various factors that have to be considered in making such a choice.

The spectrograph had to be small and light enough to be mounted in a truck, and yet have adequate dispersion and resolution to permit the analysis of the complex spectra emitted by most types of geological materials. A plate factor of 7A per mm was set as the minimum dispersion that would be satisfactory. Moreover, it should permit the photographing of a range of spectrum from about 2,200 to 4,800A in a single exposure, because the sensitive lines of the majority of elements usually determined in geological materials fall in this region. Other factors considered in the selection were lightness, compactness, rigidity, and cost. Although not essential, a stigmatic mounting of the grating was considered more useful than an astigmatic one, because such a mounting permits the use of a number of effective methods of photographic photometry not possible with an astigmatic mounting, but useful in quantitative spectrographic analysis.

Spectrograph and Direct Accessories—A survey of spectrographs available commercially in 1952 revealed that no one instrument would completely meet all of the requirements considered essential for a truck-mounted instrument to be used in the analysis of geological materials. However, one instrument did meet most of the qualifications except that of wavelength coverage. This was a small, compact, fixed-position spectrograph employing a 1.5-meter replica grating in a Wadsworth stigmatic mounting, which recorded the spectral region from 2,100 to 7,800A in the first order, and 2100 to 3900A in the second order with plate factors of 10.9A per mm and 5.45A per mm, respectively. Consultations with the manufacturer of this instrument indicated his willingness to modify this item to meet our requirements. Accordingly one of these instruments containing a 15,000-line-per-inch original concave grating was supplied with a modified frame to cover the wavelength range from approximately 2,050 to 4,850A in the second order on a 20-inch length of film. To avoid the difficulty of overlapping orders, some consideration was given to obtain this wavelength coverage in the first order by using a 30,000-line-per-inch grating, but uncertain availability of 30,000-line-per-inch gratings ruled against further consideration. Also, the use of a 15,000-line-per-inch grating makes available, if needed, a wavelength range of from 4,100 to 9,700A in the first order; this permits the use of the most sensitive lines of a number of elements, notably the alkali metals, that lie in this region.

Ordinarily, however, very little of the first order spectrum is recorded when "unsensitized" photographic emulsions are used, e.g., those that are insensitive above about 5,000A. The third order spectrum up to about 3,225A is also recorded but this overlapping ordinarily causes little inconvenience.

The instrument as received proved to have an exact wave-length coverage of 2,063 to 4,837A in the second order (4,126 to 9,674A in the first order). The 15,000-line-per-inch original concave grating with which it is equipped is of good quality and gives a well-defined and clean spectrum of uniform speed throughout the second order. The manufacturer reported that when the second order spectrum was examined under an 80-power microscope, his tests indicated that it was possible to resolve the two lines of the iron 3,100 "triplet" that have a separation of 0.07A. Our own tests have shown that the instrument is also very fast, a 3-second exposure being sufficient to produce a strong spectrogram of the iron spectrum.

The spectrograph is shown in Figure 4 and the optical bench section and the control panel for the arc source are shown in Figure 5. The instrument has a compact, rigid frame of welded steel plates and a sheet steel cover. It is 67 inches long, 37 inches wide at the filmholder end, and 10 inches high. The optical bench extends out about 13 inches. The film holder is designed to use a strip of 35-mm film, 21-1/2 inches long, which is loaded through a slot at one end. A dark slide permits the holder to be removed from the spectrograph and carried to the darkroom without danger of accidental exposure of the film. The filmholder is racked manually by a handwheel located at the front of the instrument near the slit.

Direct accessories for the spectrograph, not all of which are used at any one time, include a 3-position fixed slit with slit openings of widths 10, 20, and 100 microns; 200-mm spherical quartz lens for focusing an image of the arc on the collimating mirror; 50-mm spherical quartz lens for focusing the arc on the spectrograph slit; arc stand; target and lens for aligning electrodes; electromagnetic shutter and timer; rotating sector mount with variable aperture and stepped sector discs; and filter assembly with combination 3- and 7-step rhodiumized neutral filters on quartz.

As furnished by the manufacturer the instrument could be focused by manually moving the slit assembly back and forth in a draw tube. The slit aperture could be aligned with the grating rulings by rotating the slit assembly in the draw tube. In the early stages of this project, frequent adjustment of the slit assembly was anticipated. The mounting of the slit assembly, therefore, was modified in the machine shop of the Geological Survey so that both adjustments could be accomplished accurately and reproducibly by lateral and tangential adjusting screws. The only other modification made to the spectrograph was the installation of lock-nuts on the adjusting screws of the grating and mirror holders.

Comparator-Densitometer.—As a companion unit for the appraisal and measurement of the spectrograms, a comparator-densitometer was obtained from the manufacturer of the spectrograph. This instrument is on a lower table beneath the spectrograph (Figs. 3 and 4). It projects images of the sample and master films side by side at 10-power magnification for qualitative

and semiquantitative analysis. Quantitative measurements of spectrum line density can be made by passing the projected image of a spectrum line across a fixed-width slit, beneath which is a barrier-layer photocell connected to a sensitive galvanometer. The galvanometer spot is projected onto a scale that is located directly above the projected spectra. A motor scan unit is provided for reading the transmission of the spectrum lines. Other adjustments include zero set, sensitivity and damping controls, and individual and combined film feed knobs. The top plate carrying the film stages can be rotated manually to align the spectrum with the slit. The instrument operates from the 115-volt a-c line through a voltage regulating transformer.

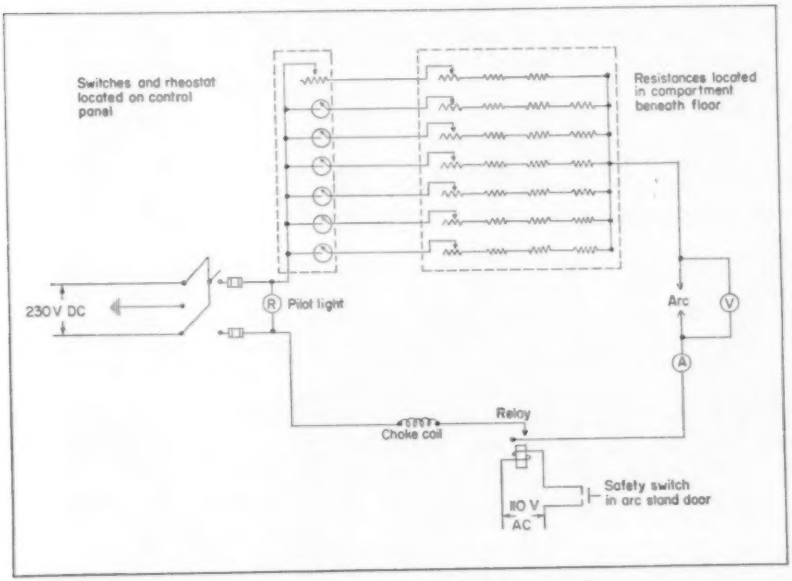

FIG. 6. Schematic circuit diagram of d-c arc source.

Arc Source Unit.—Because the laboratory would be used primarily to analyze geological materials, a source unit offering a wide variety of excitation conditions was not considered essential. The d-c arc has long been used by spectrographers for the analysis of rocks, minerals, soils, and related materials, and such a source was chosen for the truck laboratory because of its simplicity, high concentrational sensitivity, adaptability, and low cost.

Because the dimensions of the limited space available for the excitation unit dictated the size of the unit, no satisfactory commercially available model was found. Most commercial units are complex and costly units that offer a variety of excitation conditions not needed in this application. Therefore, a custom-built d-c source unit was constructed to fit in the available space.

This plan seemed to be more satisfactory than that of modifying the design of the laboratory to accommodate a commercial source unit.

A schematic circuit diagram of this source unit is shown in Figure 6. The basic design of this circuit is well known and modifications of it are in use in a number of spectrographic laboratories. Essentially, the circuit consists of an arc operated by current from a 230-volt d-c generator with ballast resistances in series with the arc to achieve stability and provide a means of current control. A large choke coil is placed in series with the arc to dampen incipient oscillations caused by rapidly changing conditions in the arc column. This circuit provides from 3 to 21 amperes of current in steps of 3 amperes. Current selection is controlled by changing the number of calibrated resistor banks in parallel with each other, the changes being accomplished by means of 6 toggle switches located on the control panel below the optical bench (Fig. 5). In one of the resistor banks, a rheostat located behind the control panel has been substituted for one of the fixed resistances to provide a quick adjustment of the current, if required. This rheostat can also be used to provide a limited amount of manual current control during the arcing of a sample. This type of circuit has the advantage of allowing the current to be increased or decreased during the burning of a sample, because switching resistor banks in or out of the arcing circuit does not involve lines carrying more than about 3 amperes.

The 7 resistor banks are mounted in a small compartment located beneath the truck floor. Such a location was chosen to eliminate from the laboratory the considerable amount of heat emitted by these resistances. When the source unit is used, the door to this compartment is open to provide ventilation. The experience to date has been that this compartment does not become unduly hot and forced ventilation has not been necessary.

The voltmeter and ammeter are on the rear wall behind the optical bench, directly above the target screen used to maintain the alignment of the electrodes during arcing. Prior to starting the arc the voltmeter records the open line voltage of the d-c circuit, and after the arc is ignited, the voltage drop across the arc gap. The ammeter records the current flowing in the arc circuit. The location of these two meters behind and above the target screen facilitates the correlation of the arc current and the voltage drop across the arc gap with the image of the arc on the screen.

A safety circuit is provided that prevents the electrode holders from being energized until the arc stand door is closed. Thus, should the arc be extinguished inadvertently during a burn and the operator fail to disconnect the main switch before attempting to change electrodes, he is protected from electrical shock because the opening of the arc stand door cuts off the power from the electrode holders. A large red pilot light at eye level on the wall behind the optical bench is another safety device to indicate if the main switch is in the "on" position.

Darkroom and Film Processing Equipment.—The darkroom in the right forward corner is separated from the remainder of the interior by two layers of black plasticized material. Wooden baffles provide a light seal at the top and bottom, and one enters the darkroom through overlapping drapes of the

plastic material. One corner of the spectrograph projects into the darkroom through an opening in the curtains; the junction of the material with the spectrograph is sealed by strips of cloth tape. All development and film loading can be carried out in complete darkness.

The film processing unit is a completely self-contained bench model which has tanks for developer, fixer, short stop and wash water, and a heated compartment for forced-air drying of film. The tanks containing these solutions are immersed in a thermostatically controlled water bath that maintains the temperature of the solutions at the selected value. The temperature of this bath is controlled by regulating the inflow of cold water and by adjustment of the thermostat that controls the heating element. The unit contains a built-in timeclock that rings a bell at the end of the selected development period; it also has an automatic timer to control the drying of the film in the forced-air compartment. In the standard model of this film processor, film is washed by means of continuously running tap water. Because of the limited supply of water in the truck, the processor used in the truck was modified by the manufacturer to provide the wash water tank with agitation similar to that in the developer and fixer tanks. The fixed film is washed with several changes of water.

Cold water for the constant-temperature bath in the film processor is supplied by a combination water cooler-refrigerator. To conserve water, the cold water from the reservoir in the water cooler is pumped to the film processor by a small centrifugal pump, and returns to the water cooler by gravity flow. The top of the unit is covered by a metal plate, which is used as a table for loading the film holders. The refrigerated compartment is used for the storage of film. Insulation and ice cubes frozen in the freezing coils of this section during use help to maintain a cool temperature when the unit is shut down at night. The film is protected from melt water from the freezing coils and changes in humidity by being stored in tight metal cans, but it has been the practice to remove new film from refrigeration at least a day before use to permit the emulsion to reach equilibrium with the temperature and humidity of the field station.

Water for film washing and other uses is supplied to the faucet by a second centrifugal pump from the large water tank over the left wheel-well. Waste water is discharged directly to the ground through a pipe and trap beneath the cup sink.

Considerable storage space for supplies of chemicals and solutions is available beneath the processor bench. A supply of demineralized water for making up developer solutions is also carried here in plastic bottles.

Accessories.—A tank-type vacuum cleaner is mounted on the right wheel-well beneath the densitometer table. Its main use is to clean the electrode holders and interior of the arc stand between samples to prevent the condensed residue from one sample contaminating the next sample.

An electrode shaper is available for the fabrication of special electrode shapes desired for specialized applications, but is not otherwise used, because preformed electrodes are satisfactory for the routine methods.

A 50-mg torsion balance is used to weigh out the requisite amounts of

sample and graphite powder used in the routine analytical scheme. A specially fabricated thin sheet aluminum weighing pan facilitates the transfer of this charge to the cavity of a preformed electrode.

Other accessories carried as a part of the laboratory equipment include a drying oven, electric heater, mortar grinder, agate and ceramic mortars of various sizes, and a ceramic buckboard and muller.

PROTECTION OF EQUIPMENT AGAINST VIBRATION AND SHOCK

As suggested earlier, much thought was given to the problem of how to mount and arrange the laboratory equipment in the truck so that it would be protected against road shock and vibration that might damage it or throw it out of adjustment. Protection of the spectrograph and densitometer was the most critical. The scheme finally adopted for both these items was to mount them on rubber shock mounts of suitable capacity and design. The flexible mountings used, in addition to securing both items to their respective tables, not only protect them against steady vibration but also cushion severe shock loads. This system has proven fully satisfactory. One of the spectrograph mounts can be seen in Figure 5.

The combination water cooler-refrigerator is bolted directly to the floor in accordance with the recommendation of the manufacturer who believed that the regular motor shock mounts would be sufficient to protect the unit.

When the truck is set up in a new location, a screw-type house jack is placed under each corner of the truck body to lift the laboratory off the truck springs and thus minimize the vibration caused by movement of personnel in the truck that might interfere with weighing operations or measurements with the densitometer. These jacks are also used to level the body of the truck.

All of the drawers and storage compartments are lined with sponge rubber or felt to protect the supplies stored in them.

Items of equipment, such as the electric oven and film processor, that are not susceptible to shock or vibration damage are fastened directly to the floor or tables.

REQUIRED PREPARATION OF LABORATORY FOR TRANSIT OR OPERATION

Because most of the laboratory equipment is built in or permanently secured to tables and benches, little time is required to prepare the laboratory for transit. All of the large movable items such as chairs, file drawers, etc., are secured to the walls by springs, and the smaller miscellaneous items are stored in drawers or storage compartments. All drawers and sliding doors are secured with fasteners to prevent vibrating open when the truck is moving on the highway. The spectrograph and densitometer require very little attention to prepare them for movement. For the spectrograph, cover plates are placed over the openings to the grating and mirror and sealed to the housing with masking tape to prevent dust from collecting on the surfaces of the mirror and grating. The junction between the top cover plate and the main body of the spectrograph is sealed with tape, and a plastic cover is fitted over the 3-position slit. The arcing stand, which can be removed easily from

the optical bench by releasing one screw and disconnecting 3 electrical plugs, is also removed for transit. The densitometer is prepared by clamping the galvanometer suspension and fastening the film carrying stages down with straps. In the darkroom, solutions are removed from the film processor and stored in bottles, and the water level in the water cooler reservoir is lowered so that the motion of the truck will not cause any spillage. Cables from the mobile electrical trailer are disconnected and stored on cable reels on the trailer. The jacks are removed from under the truck and are stored on the trailer also.

Preparing the unit for use is essentially the reverse of the above operations. When the laboratory is operating, the power trailer is located about 100 feet forward, or to one side of the truck. This arrangement minimizes both the vibration and noise that would result if this unit were mounted in or near the truck.

RESULT OF FIELD TRIALS

In June 1955 the truck-mounted laboratory was driven 525 miles to the East Tintic mining district near Eureka, Utah, for a tryout period of one week. Although there was a Geological Survey geochemical exploration drilling project under way in the area that welcomed the rapid on-the-spot analytical data provided by the truck, the main purpose of this trip was to find out how well the spectrographic equipment would remain in adjustment after being driven a considerable distance over roads. Secondary aims included the determination of the time required to prepare the laboratory for transit and use at a field location, possible improvements in type and arrangement of equipment, and optimum size of crew. Although it would have been possible to obtain the answers to some of these questions merely by driving a certain distance in the vicinity of Denver and then returning, such an arrangement could not substitute for taking the unit to a field project and operating in a location where well-equipped repair facilities were not readily available.

Before leaving the Denver area a careful check was made of the condition and alignment of the spectrograph and densitometer optics. A number of alignment and focus films were taken which could be used after the laboratory had been established at the field location as a photographic check on any possible change in the optical alignment and appearance of the spectrum as a result of the trip. In addition, visual checks and measurements were made of such things as the focus of the arc on the collimating mirror, the alignment between the mirror and grating, and the location of the spectrum in the film aperture.

The trip of 525 miles to the field location required approximately 20 hours. During the trip, both to and from the field area, no effort was made to pamper the equipment because of road conditions; such equipment units, if they were at all feasible, should be capable of withstanding severe usage.

Upon arrival in the East Tintic district, the unit was set up in an isolated area completely away from any urban facilities. In less than two hours the laboratory was ready for operation. Inspection of the equipment showed that all of it had survived the trip undamaged and appeared to be in good

operating condition. Checks were made of the spectrograph and densitometer optics by comparing the measurements with those taken in Denver prior to leaving. These showed that the grating holder had rotated slightly during the trip so that one end of the spectrum in the film holder was tilted approximately 1 mm over a 20-inch strip of film. However, because this amount of rotation caused no visible change in the sharpness of the spectrum lines, no corrective adjustment was considered necessary.

During the field trials the laboratory was operated by a crew of four. Although there are four working locations in the truck, some time was lost with this size crew, and we believe that a crew of two or three would be the most efficient and result in the greatest number of determinations per man-day.

In the darkroom, no trouble was experienced in the processing of films. The time required for the processor to reach the desired temperature of development (20° C) generally averaged about 1 hour and was of course dependent on the room temperature of the laboratory at the time the power was switched on in the morning. More time was required when the initial temperature of the water bath was over 20° C than vice versa, because of the longer time required to cool the water in the reservoir. The flow of cold water was adjusted so that the heater in the processor operated only for a short period every 3 to 4 minutes. Once the film processor had reached the desired temperature and equilibrium was established, temperature regulation was such that the temperature of the processing solutions did not vary more than 1/2 degree from the selected temperature. After a period of use, some adjustment of the cold water flow was usually required, owing to an increase in the room temperature of the darkroom. During the tryout period the weather conditions ranged from cold and wet with maximum daytime temperatures in the low 40° Fs during the first few days, to hot and dry with maximum temperatures in the high 80° Fs at the end of the period. During the colder days the small electric heater was used to heat the interior of the truck. Conversely, the air conditioner was used during hot weather to make the interior reasonably comfortable even during the hottest part of the day. Cooling was most effective in the forward section of the truck, and less effective at the back, especially when samples were being arced.

After the truck laboratory had returned to the Denver headquarters, a thorough inspection of the laboratory showed that all components had survived the return trip in good adjustment. The grating had not rotated any further and the quality of the spectrum was as good as before the field excursion.

In November 1955 the unit was first used to supply routine on-the-spot analytical data in support of a geochemical exploration field project in south Texas. On this trip the truck was driven over 3,000 miles. The usual checks of optical alignment were made both after the arrival at the site of field operations and after the return to Denver. No change in the adjustment of the various spectrographic components was detected. This confirmed the findings of the field trial in Utah, that spectrographic equipment and accessories, when properly mounted and protected, can stand the rigors of highway travel.

The laboratory was initially set up for the determination of 33 elements: antimony, arsenic, barium, beryllium, bismuth, boron, cadmium, calcium, chromium, cobalt, copper, gallium, germanium, indium, lanthanum, lead, magnesium, manganese, molybdenum, nickel, niobium, scandium, silver, strontium, tantalum, thallium, tin, titanium, uranium, vanadium, yttrium, zinc, and zirconium. With the proper standards, the laboratory could be used to determine 35 additional elements that are capable of analysis in the d-c arc, e.g., aluminum, cerium, cesium, dysprosium, erbium, europium, fluorine, gadolinium, gold, hafnium, holmium, iridium, iron, lithium, lutecium, neodymium, osmium, palladium, phosphorus, platinum, potassium, praseodymium, rhenium, rhodium, rubidium, ruthenium, samarium, silicon, sodium, tellurium, terbium, thorium, thulium, tungsten, and ytterbium. With few exceptions, the sensitivity obtained on the truck-mounted spectrograph compares favorably with that obtained on large stationary models in the laboratory. For example, among the elements easily determined spectrographically, as little as 0.0001 percent copper and 0.0001 percent silver can be determined in the truck laboratory. Among the elements for which spectrographic sensitivity is low, about 0.05 percent each of arsenic or zinc is the lower limit that can be determined.

Although a reliable figure on the productivity of this laboratory will be obtained only after it has been completely staffed and this staff has completed a number of routine assignments, the output of the laboratory while in the field in Texas can be used as a rough approximation. There, one man made 2832 semiquantitative determinations on 354 samples in 12 days, an average of 236 determinations per day.

Samples collected by the field party were brought to the laboratory at the completion of a day's sampling and selected suites, designated by the party chief, were placed in an electric oven to dry. The next day these samples were prepared by one of the samplers in the party and delivered to the spectrographer. Analytical results were routinely available to the party chief less than 48 hours after collection. Smaller groups of high priority samples could, of course, be analyzed in a much shorter time if desired. The availability of this on-the-spot data facilitated greatly the progress of the geochemical field investigations.

Indicated Improvements.—The design of any new piece of equipment can always be improved after it has been in use for a time. The mobile spectrographic laboratory proved to be no exception because a number of improvements, minor in nature, were indicated as a result of the field trials.

The darkroom part of the laboratory becomes excessively hot during periods of warm weather. This is mainly because this section is separated from the remainder of the interior by a curtain and contains a source of heat in the form of the heat exchanger of the refrigerator. The poor dissipation of heat from the exchanger, because of the close quarters, not only makes it uncomfortable for the personnel using the darkroom, but taxes the refrigerating capacity of the water cooler; therefore, plans are being made to provide some ventilation of the darkroom.

The electrical plants on the trailer are sturdy units and give no trouble.

We believe, however, that the substitution of a 5-kw model for the 3.5-kw, 110-volt, a-c plant, would be a desirable change in the design of any future truck laboratory because the power demand on the latter plant occasionally equals the capacity.

Although it is possible to do satisfactory visual matching of unknown and standard spectra using a 10-power magnification, we believe a somewhat greater magnification would result in less eye strain and permit more precise matching. Also, the experience gained during the tryout period indicates that the comparator-densitometer will be the limiting factor in controlling productivity under most conditions. Therefore, some consideration is being given to the desirability of obtaining a small portable comparator for visual matching only; the latter could be set up and used when the backlog on the regular instrument is too great.

A second torsion balance for weighing out samples would be a desirable addition because the semiquantitative spectrographic method employed for routine analyses involves the weighing out of a 10-mg sample, mixing this with 20 mg of graphite powder, and then loading the resulting mixture into a graphite electrode. The second balance would result in a more efficient utilization of the crew during the first day or two of a field assignment when the major tasks involve preparation of standard films in addition to preparation of unknowns.

SUMMARY

The feasibility of transporting and using spectrographic equipment in a truck-mounted laboratory has been demonstrated, although many of the fields of maximum usefulness of such laboratories are still to be determined. Because of the large number of elements that can be determined from a single spectrogram, the truck-mounted laboratory is especially useful in the early stages of a geochemical exploration project to determine diagnostic suites of elements; later, it may still be more efficient for guiding the day-to-day sampling than wet analytical methods, if many different elements are to be determined. We believe that here is another tool for geochemical exploration which, under certain conditions, should provide the fastest and most economical way of obtaining needed data on the dispersion of traces of metals in geological materials.

U. S. Geological Survey,
Denver, Colorado,
Nov. 23, 1956

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SCIENTIFIC COMMUNICATIONS

THE COSMIC ORIGIN OF METALLOGENETIC PROVINCES

A. C. SKERL

It is postulated that at irregular and increasing intervals during the history of the earth there have been brief periods of intense bombardment by meteorites originating from the break-up of cosmic bodies of various composition.

The target areas struck by metallic meteorites have developed into metallogenetic provinces either directly as sedimentary deposits or indirectly through assimilation, granitization and subsequent hydrothermal action.

INTRODUCTION

A study of a recent account of the present knowledge and theories concerning the origin of the earth by W. M. Smart gives the impression that the cosmologists are becoming more confused because each new physical fact that is discovered leads to the destruction or at least modification of one theory and the proposal of several others (1). The inherent nature of the problem is, of course, the reason for the perplexity, namely to find a satisfactory theory that will account for all known facts about the earth moved back in time some 4,000 million years or more.

As the cosmologists surround the problem with increasing amounts of higher mathematics, nuclear physics and newer theories of astronomy the geologist finds himself a puzzled onlooker. He will find however that it is generally accepted that:

1. the earth passed through a wholly molten stage early in its existence.
2. meteorites are the evidence that a cataclysm has shattered at least one solar body into fragments.

The following discussion is largely based on these two statements.

THE ORIGINAL EARTH

It is customary to consider that the original molten mass solidified into a sphere of rock with uniformly layered shells around a central core according to the densities and freezing points of the mineral assemblages stable under the prevailing conditions.

Subsequent localized melting of parts of the interior resulted in the phenomenon of intrusive rocks and their associated mineral deposits. With uniform shells of rocks these intrusions and the mineral deposits would be ex-

pected to have a regularity of composition and distribution but actually they are scattered as groups in irregular and usually dissimilar areas. Hence the statement of Bateman "The fact of metallogenetic provinces is undisputable but their cause is a problem" (2). Again no two mineral deposits are alike and adjacent mineral districts can be surprisingly different.

It is suggested that this heterogeneity of ore deposits and of their metallogenetic provinces is primarily due to a cosmic origin for much of the metallic material. This is in direct contrast to the established theory that the metals are derived ultimately from deep below the crust.

METEORITES

The composition of meteorites varies from purely stone to purely metal (mostly iron with minor nickel) suggesting that they had a common source. They probably arose from the cataclysmic disintegration of a planetary body that for some reason became unstable. They are generally associated with the passage of meteors whose paths indicate that they belong to the solar system. Astronomers allow that a few may have originated outside of the solar system.

Until recently it was thought that meteorites such as we know today have been falling on the earth for only a short period in geological time beginning in the late Tertiary. V. E. Barnes now reports evidence for the type of meteorites known as tektites occurring in the Eocene of Texas (3).

The total amount of fragments from any given body flying through the solar system is gradually reduced as they are caught by the gravitational field of a planet or the sun and so absorbed. Thus the astronomers report that the comets whose paths bring them back near the earth at definite intervals are much smaller at each return.

Spectacular meteorites weighing up to several tons have been described and some of them are exhibited in museums. The meteor crater in Arizona that is 4,000 feet across and 500 feet deep must have been formed by an extremely large meteorite (millions of tons are suggested by one authority) (4).

Far more numerous however are those in the form of showers that are known to have covered as much as two million square miles as in the case of the australites which are small disk-shaped meteorites of glassy material that are found over a large portion of Australia.

Another example, with which the writer is familiar, is the large area of southern Luzon in the Philippines in which spherical tectites up to four inches in diameter are found in the recent gravels. They consist of a black glass similar to, but distinct from, obsidian in composition. They are regarded as meteorites by Dr. Otley Beyer of Manila who is an authority on their occurrence (5). The local people call them the "mother of gold" since they are often found in the gold placer deposits.

In remote geological time there may well have been a number of planetary bodies of various compositions that became unstable and disintegrated. It is therefore conceivable that since the earth became a separate entity there have been a number of periods of meteoric bombardment of varying intensity and probably on a more tremendous scale in the very early periods.

The numerous and extensive craters on the moon are regarded as possible

evidence of the enormous bombardment with meteorites that has taken place since the moon began to solidify some 4,000 million years ago. They have been preserved by the lack of an atmosphere to support the processes of weathering.

FORMATION OF METALLOGENETIC PROVINCES

During the early molten stage of the earth it is imagined that it would rapidly engulf and then incorporate the additions from outer space but as the viscosity increased with cooling so the dispersal would be less and the target areas would become enriched by the extraneous material. The composition of this introduced material would depend on that of the original body that became disintegrated, which could have been any of the elements in any proportions and combinations. At a later stage when solidification was sufficient for the successive processes of weathering, transportation, sedimentation, and deep burial any foreign material would either be dispersed or concentrated according to its nature and where it fell. Thus in a water covered area it would be concentrated as a layer in the sediments forming at that time.

Eventually deep enough burial would lead to granitization and the subsequent formation of the mineral provinces that we know today. Areas that escaped granitization and eventually re-emerged could become the so-called sedimentary ore deposits whose origin is so controversial.

EXAMPLES OF SEDIMENTARY ORE DEPOSITS

Rhodesia and Belgium Congo.—One remarkable fact about the Rhodesian-Congo copper belt is the variety of sedimentary rock types that are the hosts for the ore deposits. Another is the very limited time range represented by each host rock and the possibility that they belong to the same time range.

During the past few years there has grown the conviction amongst Rhodesian geologists that the Rhodesian copper ores are syngenetic in origin but the manner in which they were formed is a subject of much controversy.

The widespread nature of the ore deposits contrasted with their very limited vertical range and, therefore, the brief period of time for formation is a difficult situation to envisage by ordinary weathering processes.

Garlick (6) has propounded a sensitive balance of special sedimentary conditions whereby the copper and other metals derived from the weathering of an old land surface were precipitated from solution by bacterial action. That these extraordinary conditions could be so widespread and yet limited to such a short period of time is difficult to concede.

If a source outside of the earth is assumed in the form of one or more large meteoric showers then the limited time factor and variety of host rock can be reconciled.

Much of the material would be in the form of dust and over water areas would become incorporated directly in the sediments. Much of that which fell near water would soon be swept into the sedimentary basin—in large amounts for a limited period and then the remainder would be rapidly diluted by the predominating rock debris.

The sulfide nature of the present minerals could be explained as either: a), the original form of the metals in the meteorites that was not changed on entering the earth's atmosphere because it lacked oxygen in Precambrian times or, b), reduction by sulfur present in the sediments.

Colorado Plateau.—Most of the uranium ore deposits of the Colorado Plateau are characterized by being localized in old stream channels. In their descriptions of the deposits writers have suggested various sources for the uranium—from detrital to hydrothermal but with little conviction.

A meteoric source would provide heavy particles that would accumulate in the stream channels but would disintegrate on the adjacent temporary land areas of these lacustrine deposits.

Blind River.—The unexplained but presumably significant alignment of the uraniferous areas in the Blind River field of Ontario, in a belt that crosses the fold pattern, leaving large areas of otherwise favorable conglomerate host rock non-productive, could be due to an original shower of radioactive meteoric dust at the time of formation of the conglomerate.

CONCLUSION

The concept that meteoric showers in early geologic time have resulted in the metallogenetic provinces of today is at first startling but it does seem feasible. The theory offers an explanation for the haphazard distribution of mineral areas and for the controversial but important non-ferrous sedimentary ore deposits.

1758 Western Parkway,
Vancouver 8, B. C.,
Jan. 18, 1957

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DISCUSSIONS

A CRITICAL EVALUATION OF THE CLASSIFICATION OF ORE DEPOSITS OF MAGMATIC AFFILIATIONS

Sir: The following comments are inspired by A. D. Mutch's article in the November issue of ECONOMIC GEOLOGY.

From time to time the legitimate dissatisfactions inseparable from the Lindgren classification come to a head, and we are presented with a statement of its defects and some suggestions that are different, but rarely new. Some ideas, like those of J. S. Brown, present novel assumptions as to the genesis of ores; these serve the useful purpose of jarring our complacency. Others represent a resigned return to morphological description, and it is in this group that Mutch's article falls.

Field geologists will recognize that replacement and fracture fillings, lenses and stockworks, schlieren and saddle reefs, are not basic criteria for classification, but rather the end-products of more fundamental variables; pressure, temperature, and physico-chemical environment. The fact that the position of a deposit in a genetic classification is uncertain, or ambiguous, does not invalidate the classification itself. Lindgren viewed the T-P classification not as a finished work, but as an ideal to strive for; one subject to constant modification by specific local evidence, and especially by field geology. Those critics who read Lindgren in the original, as I am sure Mr. Mutch has done, will frequently find their most valid objections directed not at Lindgren, but at the superstructure presented by Professor Graton. Most critics of Lindgren have had less field experience of mineral deposits than he, and those few whose knowledge of physics and chemistry surpasses Lindgren's have inevitably done even less field study. Mr. Mutch is to be congratulated on his frank admission of these limitations.

Mutch's eight criteria, several of which are further divisible, may prove useful as a graphic shorthand, but to envisage a classification based on eight variables is to regress and invite confusion. One may justifiably criticize the elusive T and P for varying within a single deposit, but, as they vary, six of Mutch's criteria vary too, proliferating the chaos rather than eliminating it. Inversions of logic ensue, such as "the temperature of formation of a deposit was dependent on the economic mineral that was present" (p. 677), as much as to say the product is the cause of the agent.

How are we to interpret the desire for a "less infallible descriptive term" (same page)?

> "Sometimes these cogitations still amaze
> The troubled midnight and the noon's repose"
> —T. S. ELIOT

Mr. Mutch wishes to free his data-sheets from theoretical considerations, yet he leans heavily on "magmatic affiliation" and "the standard paragenesis"

two terms so loaded with genetic implications as to have a mystical value for many geologists. One may hope for a measure of certainty in determining the temperature and pressure of formation of a granitic rock, but it takes a wealth of inexperience to label it magmatic or metamorphic with complete assurance.

By all means let us guard our non-genetic terms unsullied, for factual descriptions, but not be misled into thinking that a cogent or meaningful classification can be based on them.

ROBERT GREENWOOD

UNIVERSITY OF HOUSTON,
HOUSTON, TEXAS,
Dec. 1, 1956

PROFESSIONAL ETHICS AND CONDUCT FOR ECONOMIC GEOLOGISTS

The vastly expanded search for minerals, with a resultant increase in the ranks of economic geologists since World War II, coupled with the mushroom growth of uranium prospecting and development which involved the formation of scores of uranium companies and the offering of innumerable shares for public consumption combined to pose problems of ethical conduct on a scale not hereto encountered in the realm of Economic Geology. In view of this, in 1954 a committee was appointed by the then president of the Society of Economic Geologists for the purpose of studying professional ethics and conduct to determine the desirability of a permanent committee on ethics, as well as a formal written code for the society.

This committee, under the chairmanship of Harold Bannerman, consisted of R. N. Hunt, Ira Joralemon, William James, Hugh McKinstry and Olaf Rove. They served with commendable effort and efficiency. Their report was published in ECONOMIC GEOLOGY, No. 5, in 1955. Briefly, they concluded that a permanent committee on ethics was unnecessary; also that spelled out rules of conduct would prove ineffectual. Instead, they recommended that the Council request certain members of the society to contribute articles on various phases of ethical conduct for publication in ECONOMIC GEOLOGY. This paper is in response to such a request.

The founders of the Society wisely included the worthy generality in Article III of the constitution that one of the objects of the society was to "protect and maintain a high professional and ethical standard in the profession." In the relatively uncomplicated times when the society was formed, this stipulation doubtless sufficed—especially in view of the impeccable standing enjoyed by the founding fathers themselves. However, in these more hectic days it is timely to appraise the situation with an eye to bringing back into focus the high principles that we in the society are proud to claim as a heritage handed down from and established by our distinguished predecessors.

A wise and seasoned general practitioner recently remarked that to the pat saying "nothing is sure but death and taxes" should be added the word change. The observation is especially apt in appraising the expansion in economic geology since World War II. The ever increasing need for more

of almost every kind of mineral, coupled with the fact that obvious deposits having been found requires search at depth for hidden ores, has brought about the greatest upsurge in exploration in history. As a natural corollary, the ranks of professional geologists, as well as their activity, have increased to unprecedented scope. Inevitably, new technical problems are posed, and inevitably the old human problems are magnified. It is these problems of human relationships which are, I believe, more important than those involving scientific techniques. We Americans, especially, are adept at overcoming technological barriers. It is the other realm that merits our attention.

While it may rightly be said that morals and ethics are somewhat dependent on geography, nevertheless, peoples of all places must learn to live together harmoniously or else they perish either by means of violence or emotionally. Granted, then, that harmonious relationships must be a universal goal, it follows that all peoples must establish a basis for these right relationships, which in turn depends on appropriate conduct based on an acceptable ethical standard. In our profession, such ethical standards of conduct, as they pertain to economic geology, may logically be grouped according to intra-professional, employer and public relationships.

Inter-professionally, the complicated nature of today's ore-finding techniques requires much teamwork, which demands that professional geologists subordinate to an extent their personal interests for the welfare of the group. In the budding era of mining geology, it was often customary for an erudite pundit (necessarily of strong personality) to expound his views on the worth of a property with vigor and with such assurance that these views were taken as gospel. In those days there were many Chiefs, but few Indians. Now it behooves us to have many Indians but few Chiefs. Today, prima donnas cannot be tolerated anywhere along the complicated chain of exploration, involving as it does teams beginning with the initial idea, the acquisition of the land, photogrammetry, the geological survey, photo-interpretative work, geochemical and geophysical surveys, climaxed by the final putting together of all scientific data by a coordinated team, the members of which must contribute in a spirit of cooperation if successful and rewarding results are to be obtained.

In private companies, government agencies, or in the consulting field, the best policy to follow is the golden rule. As applied to economic geologists, it might be well to mention specifically three commandments the transgression of any one of which goes far towards lowering the standards of the profession both internally and externally. These are: thou shalt not gossip about thy colleagues, thou shalt not belittle thy colleagues, nor shalt thou underbid thy colleague. The first two admonitions need no amplification. In the case of the third, some explanation is in order. While it is conceded universally that it is unethical to underbid a professional competitor in an effort to get a job of routine consulting, it is quite in order to submit a proposal in response to a request for a large-scale operation, which proposal is on a recognized competitive basis. Also, consultants overseas in soft currency countries will perforce offer services at lower rates generally than those quoted from hard money areas.

In mentioning fees, these appear, for consulting geologists, to fall into three categories. First, are those cloistered in our ivory towers. Their

competence is unquestioned and they are essential both to the profession and the industry. However, their cost of doing business, so to speak, is negligible. They have few overhead charges, such as rent, permanent staff, and no unproductive time between jobs.

In the second category are some of our finest contributors to the profession, namely, the single consultant, or the small partnership, men who are, one might say, self-employed. They may or may not have offices, but in any case their cost of doing business is relatively low compared to the third class, namely, the larger firms or corporations who must maintain a permanent staff of experienced personnel in order to be able to meet the demands of industry and governments for group service by highly trained technicians and scientists. Experienced staff men come high. They must be paid an annual salary which is competitive with that in industry. Yet, the maximum amount of working time for such an individual is less than 270 days per annum. In addition must be considered the cost of insurance, pensions, and possibly some prerequisits so that in sum and substance a consulting firm these days to be prudent must add from 80 to 150 percent of a technical man's salary for overhead—the cost of doing, or perhaps remaining in business in order to make available for a vital demand, teams of trained consultants.

Those in category one may feel justified in charging a per diem fee of "X" dollars. Those in the second class may have to add "Y" dollars to take care of running expenses, while those in the third class are constrained to figure their fee at "X" plus "Y," plus "Z," the latter being the cost of doing business.

As for the ethics of contingent interests, in these days of high taxes we are forced to take a second look at a practice which was once not exactly condoned in the United States, although our friends north of the border have benefited abundantly along these lines over the years.

The potential flaw, as I see it, which stems from a contingent reward, lies in the temptation to spend more of the principal's money on a questionable prospect than might otherwise be the case. The advantage lies in greater incentive and effort on the part of the explorer coupled with a larger gain if successful. The ethical answer should be an arrangement whereby the employed party forgoes part or all of his regular fee, thereby placing his interests in parallel with his principals. Also, in cases of contingent interests all those concerned are entitled to a full disclose of the facts of the arrangement.

In the field of employer or client relationships, honesty and integrity with one's employer go without saying. Loyalty to the employer is also essential not only during the term of the contract but afterward as well. Perhaps it is even more important afterwards than it is during the term of the contract. A professional geologist must consider that the work he does for a client or an employer belongs to the employer. The geologist, then, is a trustee in the matter of the knowledge which he has gained for the person who hired him. He has no right to divulge this knowledge either during the course of his employment or later without the consent of those who employed him during the time of his investigation. Even new ideas that he may develop about the district in which he is working do not belong to the geologist, but rather to the person by whom he is employed. It is unethical, then, to hold back on an

idea until one has ceased employment for a particular client or employer in order to bring this idea forth later for one's own profit.

Loyalty to a client, in fact, goes all the way except where dishonesty enters into the picture. No professional geologist is required to lower either his standards of ethics, or his own personal moral code, in order to satisfy the dishonest demands of an employer or a client.

With regard to the public, it may well be said that these days the public not only deserves, but it expects, service. As herein defined, service means honest, useful conduct to others. The days are over when an economic geologist fulfills his obligations merely by adding his name to a report which merely gives a description of the geology along with a non-committal portion on the economic possibilities. The economic geologist now has the same position of stewardship which Herbert Hoover, in 1909, ascribed only to the mining engineer. His duty now transcends not only the factual reporting on quantity and quality of ore, known and potential, but he is obliged often, in the public interest, to state his appraisal of the economics of the enterprise under consideration.

Nowhere has this more implication than during the uranium boom and bubble which fortunately terminated about the time of the Geneva Conference, giving place to a more sound climate for appraisal. The shenanigans which transpired in the early boom days were unfortunately not confined to the non-professional element, whereas the public, as well as the legitimate investment concerns, had unprecedented need of sound advice and mature counsel. Notwithstanding, and despite the yeoman service done by a handful of honest professional stalwarts, to paraphrase a great Englishman, it may be fairly stated that never in the history of our profession have so many, clients and public, been so shabbily treated by so few self-styled uranium experts. The early history of the plateau boom constitutes, on the whole, a rather sorry chapter in our professional annals.

Considering the complexity of modern-day exploration, involving as it does heavy investment in equipment in the case of photogrammetry, aerial geophysical work and photogeology, it must be conceded that dignified advertising in these enterprises is consistent with good taste. It is, in fact, an essential of the free enterprise system for it stimulates honest competition with a resulting increase in efficiency.

Perhaps the dictates of good taste would limit advertising to those who have large investments in equipment as against those who furnish purely professional services. In the latter cases, the usual card in professional publications should suffice. Certainly, in these days of prosperity, most professional men have more work than they can handle. Our problems are not those of advertising.

In concluding these rather loosely put together thoughts on ethics and conduct, I again wish to commend the work of the committee which studied the subject so thoroughly, and to go on record as being in conformity with their conclusions and recommendations.

Any attempt to spell out in detail a written code of ethics and conduct for an economic geologist would prove burdensome, indeed, and its ef-

fectiveness would be questionable. Put in simplest terms, each one of us should evolve a code for his own personal ethics and conduct. Having once set up a credo that is above reproach, then we can do no better than to follow the admonition of Polonius—"above all, to thine own self be true and it must follow as the night the day, thou canst not then be false to any man."

D. M. DAVIDSON

MINNEAPOLIS, MINNESOTA,
Dec. 17, 1956

THE ZONAL THEORY OF ORE DEPOSITS

Sir: I have read with keen interest the paper by Charles F. Park, Jr., on the zonal theory of ore deposits, published recently in the 50th Anniversary Volume of ECONOMIC GEOLOGY.

Lately I made a detailed investigation of depth changes in some ore veins of Příbram, Czechoslovakia, and a study of the literature on zoning, of which some important papers unfortunately were not known or available to Dr. Park. On the basis of my field and literature studies I feeel that I cannot agree, on the whole, with the following conclusions of Park's study:

1. Under the zonal theory, the deposition of similar minerals is explained as based upon temperature and pressure, but it also attempts to consider solubilities and other physical and chemical factors (p. 246).
2. Conditions that determine the place of deposition also determine the order of deposition. The three subjects, the depth-temperature classification, zoning, and paragenesis, are dependent upon the same chemical-physical factors. They are expressions of common phenomena . . . (p. 246).

The genetic conception of zoning, as presented by Park, by no means contradicts the conception of zoning in the sense of W. H. Emmons (4–7).

From Emmons' explanation, as well as from his Figure 1 (7) it is evident that he imagined the zoning as originating from universal solutions, which ascended without interruption [1] from individual magmas toward the regions of lower temperatures and pressures, and deposited their mineral contents successively in the form of mineral zones.

Emmons' theory has caused a double response in the geological world:

1. Many authors started from the Emmons assumption of a single ore-bearing solution, and explained the zoning either directly according to Emmons' theory, or sought to explain it by the influence of other physical-chemical factors. Most of this literature is quoted by Park.
2. Other authors recognized in their detailed geological investigation of ore regions that the zoning cannot be obtained from a single continuing ascending solution. Rather they concluded that individual "ore formations" (e.g. 15, 16) or components of an ore deposit originated from separate parts of solutions whose ascent had been separated in time and whose composition changed with time.

Some of these papers were known to Park, mainly F. Schumacher 1936, T. Watanabe 1943 and T. S. Lovering 1949, but had, however, no substantial influence on his conclusions.

[1] S. S. Smirnov (16) referred to this in a similar way.

The interrupted ascent of solutions, caused by congestion and reopening of fissures, or formation of new fissures, has been available in the literature for a long time. This information was not used for the interpretation of zoning, but it was briefly noted by F. Pošepný (14), and by Berg's (2) "rejuvenation," and his later study in 1938; in more detail this phenomenon was noted by T. B. Nolan (12) and many other authors. This interrupted ascent of solutions was also recently discussed in detail by J. Kutina (10) and G. Tischendorf (17).

F. Schumacher (15) stressed the fundamental significance of this process for the origin of individual ore formations in the Freiberg ore district, without generalization as to zoning phenomena.[2]

In opposition to the conclusions of W. H. Emmons the significance of the interrupted ascent of solutions for the origin of zoning was generalized in 1937 by S. S. Smirnov (16), reverting to some observations and conclusions of L. Hills in 1915, A. Knopf in 1924, T. B. Nolan in 1935, O. Ross in 1935, and F. Behrend in 1936.

Smirnov's opinions have been followed or discussed in several additional studies on zoning of ore deposits (about 10 papers in Russian).

During my study of vertical changes in ore veins of Příbram (especially the investigation of the Main Ethelbert Vein) I was able to find that the interrupted ascent of solutions, conditioned by reopening and refilling of fissures, played an important role in the origin of zoning (10).

In contradiction to the Russian authors, who consider zoning in Smirnov's sense (the so-called *"pulsation zoning"*) as the only kind of zoning, I think it also necessary to distinguish independently cases of zoning resulting in mineral products derived from the same solutions;[3] in other words to distinguish *monoascendent* and *polyascendent zoning* (9, 10). A more detailed proposal of a classification of zoning together with a comparison with the existing classifications will be presented by me in a separate paper.

Park's first cited conclusion might be restated as follows:

The zonal theory explains the causes of zonal distribution of mineral accumulations, as it appears, for instance, at the earth surface or with increasing depth in ore deposits.

Or, if one desired to include other factors, it might be phrased as follows:

Zonal theory is understood to be an explanation of zonal distribution of minerals, but it is necessary to include the influence of tectonic processes as well as of physical and chemical factors—both in space and time.

I refer next to the second conclusion of Park, quoted above.

One cannot agree with all three of the quoted sentences. Consider the first of them: "Conditions that determine the place of deposition also determine the order of deposition." This conclusion could be valid only for monoascendent zoning. This has been, however, only slightly considered, and it is possible that its validity may be limited. It would be useful to verify this conclusion by means of more concrete cases, based on detailed field and labora-

[2] The origin of individual formations in the Freiberg ore district, in the sense of the interrupted ascent of ore-bearing solutions, has been confirmed also by new, detailed investigations by G. Tischendorf (17).

[3] I refer to the paper of Y. A. Bilibin (3), who, however, distinguished three kinds of zoning.

tory investigation; e.g., at the moment it is not clear to me if it will hold both in cases of deposition of a monoascendent filling of an area of a blind ore vein, and in cases where tectonic movements might cause suddenly leaking of gas components from solutions, owing to which other components will crystallize from the solution.

However, this conclusion is by no means valid for cases of polyascendent zoning, which are common in nature. Here, after all, *the place of deposition was determined in the first place by tectonic conditions, whereas the sequence of deposition was by physical-chemical processes!*

I might note some concrete examples:

1. In the case of zoning described by G. M. Virovlyansky (18) galena ores were localized in the peripheral part of the area, because fissures were formed first in the center of the area and later in the periphery.[4] The place of deposition was thus determined, first of all, by tectonic conditions.

The sequence of deposition, i.e. galena being deposited as a younger component than pyrite-arsenopyrite ores (in the central part of the area), was, however, not conditioned tectonically. Solutions, from which pyrite and arsenopyrite ores originated, were no doubt of a different composition than the later solutions, which deposited galena.

2. In the Main Ethelbert-vein of Příbram, the individual mineral accretions were localized, where re-opening of the fissure took place. If the physical-chemical conditions of the solutions were correct for calcite deposition, and the fissure was re-opened at shallow depth, calcite would be deposited here as it would if re-opening of the fissure took place at a depth of almost 1½ km! The place of its origin was conditioned by tectonic factors. On the other hand, its place in the sequence of deposition (i.e. why, for example, earlier deposition of galena took place) is no doubt the result of complicated physical-chemical processes, owing to which the composition of individual parts of the solutions changed with time.

There are many similar cases.

In conclusion, I would like to express my full agreement with Dr. Park, in that both zoning and sequence of deposition of minerals must be studied simultaneously.

It should also be noted that the genetic conception of zoning, as presented by Emmons, which the conclusions of Park by no means contradict, has found no unexceptional, general recognition and active support among the chief representatives of economic geology, as may be seen, for example, from the well-known works by W. Lindgren (11) and A. M. Bateman (1, p. 274–5).

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PRAGUE, CZECHOSLOVAKIA,
Dec. 6, 1956

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[4] A penetration of pyrite- and arsenopyrite-bearing veins by galena veins has been found.

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MINERAL RESERVES AND MINERAL RESOURCES

Sir: In the Article entitled "Mineral Reserves and Mineral Resources" in Economic Geology for Nov. 1956, the authors, on behalf of a Committee of the Society of Economic Geologists, recommend for general acceptance the three category classification system for ore reserves proposed by Leith.

There have been many definitions of the three categories made and all have obvious disadvantages when applied to certain cases.

The categories are used to indicate various degrees of doubt as to the existence or grade of the ore estimated and all definitions make some attempt at defining the actual measurements required to put ore into a category. Leith's "proved or assured ore" is ore "blocked out in three dimensions by actual underground mining operations or drilling." Yet exactly the same degree of blocking out or drilling will give widely different degrees of assurance in different ore-bodies or even different parts of the same ore-body. Conversely widely different degrees of measurement will give equal assurance in different ore-bodies.

What can you do with possible ore? Can you spend money on the strength of it? If so, how much? Or must you, as the authors state, bring it into a higher category by further development before considering it an asset?

Under certain circumstances this last may be important.

The mining taxation laws in Canada, for instance, allow a three year

period without taxes for new mines. From then on profits are taxed about 50%.

The more that is known about a deposit before mining starts the greater the financial advantage that can be taken of this tax free period. Not more than a small proportion of the total ore in a mine is ever proved at the start, but it is obvious that, in Canada, probable and possible ore are an important consideration when planning the size of a mill.

A suggestion made by Dr. C. O. Swanson for classifying reserves seems to have advantages over any of the three category systems so far proposed. ("A Perspective on Mining Geology" printed in the Canadian Mining and Metallurgical Bulletin for April 1949.)

He suggests; "For reserves that are reasonably certain the tonnage estimate could be supplemented by figures giving the likely errors, which would be readily obtainable, as a rule, from the results of local experience."

He gives an example of measured ore: "500,000 tons with a likely error of 125,000, grade £ 10% metal with a standard deviation of three in the figure for the percentage."

Also he writes: "For reserves that are least certain, the existence of the ore is the critical factor in most cases and the description might be as follows:

Prospective ore 1,000,000 tons probability 0.2. The meaning of this entry would be that there are two chances in ten of finding a million tons. Reserves of this type are important in formulating long term policies and represent an expectation of future profit that may justify some current expenditure as a reasonable bet. For the specific figures given above the size of the expenditure would be related to the profit on 200,000 tons of ore."

This is similar to the practice in mine valuation of discounting ore for risk.

No attempt is made to define what degree of measurement is required for any probability. This seems realistic as every case is diffierent.

Variation in the estimated grade of ore is something very different from variation in the tonnage.

In the example of positive ore given by Swanson his tonnage might be 25% less than the figure given, and the grade given as 10% might be as low as 7%.

This grade variation might be alright if the metal was copper or lead but in the case of zinc it might be that 10% was payable but 7% not, in which case his entire positive reserve might be non-existent.

Similarly with the definition of measured ore suggested by the U. S. Bureau of Mines and the U.S.G.S., which ore can vary as much as twenty percent both in grade and tonnage. For quantity 20% seems reasonable but the grade of ore in any category can only vary a percentage well within the expected margin of profit or it may cease to be ore.

The ore in most mines is sufficiently diverse in grade that it is possible to estimate a tonnage all of which is above a grade that is considered to be the economic cut off at the time. All ore could be given with a probability figure for tonnage, but with no qualification of the grade, as in Swanson's example of prospective ore.

I take the statement "there are two chances in ten of finding a million tons" to mean either or both of the following:

1/ If five similar geological situations are tested one of them is certain to produce a million tons of ore.

or 2/ There may be as much as a million tons of ore but there is absolutely certain to be two hundred thousand.

The second points to a method for describing all ore in terms of positive ore so that valid financial estimates can be made.

An ore reserve estimate might read:

Grade	Tons	Probability		Positive ore
?	100,000	0.9		90,000
?	100,000	0.6		60,000
?	100,000	0.2		20,000
			Total	170,000 Tons.

If intelligently estimated, it seems to me that the 20,000 tons at the bottom is every bit as certain to exist as the 90,000 tons at the top so that adding them together is perfectly legitimate.

This method of estimating reserves seems to have several advantages over that recommended by Blondel and Lasky.

It is more flexible. All considerations of measurement, geology and experience can be taken into account in deciding on the probability factor as well as in estimating the tonnage. Any number of categories can be used as circumstances dictate.

Possible and probable ores are given a numerical value on which realistic financial planning can be based.

Telling the truth about ore reserves to the public is not easy.

The average non-technical investor, being ignorant of the difficulties of computing reserves, tends to consider tons of ore in a mine, like cans of beans on a shelf. If he is told that there is a million tons, that is what he expects. He, in fact, only recognizes positive ore.

If, however, he is only told of the positive ore, he is being misled by an underestimate.

If he is given the three categories he will inevitably total them up and treat them as positive. He is then possibly being misled by an over-estimate.

The only figure that means the same to both the engineer and the non-technical investor is the total of all ore suitably discounted by a probability figure.

I personally would like to see some system based on the suggestions of Dr. Swanson, recommended for general adoption by the Society, rather than the three category system of Leith.

Q. G. WHISHAW

FRESNILLO, ZAC., MEX.,
Jan. 11, 1957

URANIFEROUS MAGNETITE–HEMATITE DEPOSIT AT THE PRINCE MINE, LINCOLN COUNTY, NEW MEXICO

Sir: G. W. Walker and F. W. Osterwald have described a unique uranium deposit in a recent issue of your journal (1). I can take no exception to the article itself, since I have studied no specimens from the deposit and because the photographs are not clear enough to warrant an opinion. Figure 2a, for instance, might as well (or even probably) indicate the product of groundwater alteration of pyrrhotite, as is shown in thousands of textbook type examples, and which is not at all surprising in a pyrometasomatic deposit.

I must however comment on two points. 1. The reference to work by Meister (2) and by Berg (3) is misleading, since both reports have been completely superseded by later information, a fact that was clear to Berg before his death. The magnetite-hematite ores of Schmiedeberg, long viewed as pryometasomatic (*contact metasomatic*—Kontaktmetasomatisch—in Germany), have been definitely shown to be sedimentary ores that were thermally metamorphosed by the Riesengebirge granite. The uranium deposit in these ores is now known to be younger (probably much younger) than even the latest possible magmatic activities of the granite and it is only spatially and accidentally related to the granite. Its paragenesis, as far as the information has been developed in Germany, and which I have studied in detail, is entirely that of a normal Ni-Co-Ag-As-U vein, somewhat similar to Joachimstal, or, in view of the relative abundance of copper ore, to Great Bear Lake, or, in view of the occurrence of selenium, to the Goldfields district of Lake Athabasca. The mineralogy is complex, but the marked preponderance of Ni over Co is especially striking. Magnetite and hematite are only accidental gangue minerals spalled from the wallrock. The earlier incorrect interpretations were based on incomplete observations that noted only the tiny, thin, anastamosing veinlets, and missed the main deposit from which the veinlets branch.

2. As early as 1931, I (4) assumed, on the basis of Hüttig's work (5), the formula $3Fe(OH)_2 + O = Fe_3O_4 + 3H_2O$ for the low temperature formation of magnetite. It may however also form in other ways. Hoehne (6) and I (7) have described the very low temperature formation of magnetite in the Lothringen minettes, in which this mineral is locally very abundant.

P. RAMDOHR

HEIDELBERG, GERMANY,
Jan. 17, 1957

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REVIEWS

Die Erzmineralien und ihre Verwachsungen. 2nd edition. By PAUL RAMDOHR. Pp. 875; figs. 543. Akademie-Verlag, Berlin, 1955. Price, D.M.

Ramdohr's book on the microscopy of ores, the first edition of which was published in 1950, was a landmark in the development of a group of sciences and techniques applied to the metallic minerals. The present edition is a substantial improvement of the earlier one which was reviewed in this journal.[1] Other books dealing with the microscopy of polished sections are merely tables of certain properties and as such have been useful. The present book includes all such data and much besides.

The book is unique in the field of reference books on mineralogy in that it combines an excellent account of physical properties, etch tests, and the like with some of the results of experimental investigations dealing with geochemistry, and a critical discussion of the relation of each mineral to its associated minerals (Verwachsung). In all, some 250 metallic minerals are covered.

The book is divided into two sections: the major portion, 591 pages, dealing with systematic description and an introductory part of 219 pages which covers a wide variety of subjects from classification of ore deposits to classification of textures. In this introductory section somewhat undue space may have been given to enumeration and classification but withal it contains much material of value to advanced students of ore deposits. The reviewer feels however that a number of sections of this portion of the book fall somewhat short of a sufficiently comprehensive treatment and, if expanded, perhaps belong in a textbook of general economic geology rather than in a book primarily concerned with systematic mineralogy.

The systematic, descriptive portion of the book covers the metallic minerals with the same arrangement as the earlier edition and has been thoroughly revised. Numerous changes and additions occur in the text and outstanding is the addition of 222 new illustrations of the total of 543. The quality of these plates is remarkable: the subject matter is good, the polish of the specimens is superb, and better paper than the former edition results in a photographic record of great value.

As to some of the appendices: Ramdohr has undertaken a catalog of mineral localities with a list of major and uncommon minerals found at each. This table is useful in recording unusual minerals but is misleading in that for numerous districts even important minerals of the ores are not recorded. Of 939 localities or districts only 66 are recorded from the United States, and many of these are of trifling importance, and only 23 localities are recorded for Canada. The bibliography is very useful. In spite of new entries the total number has been reduced from 804 to 659.

This book is now in a class by itself as a source of information for the metallic minerals. No one concerned with the mineralogy of ore deposits can afford not to become well acquainted with it.

EDWARD SAMPSON

PRINCETON UNIVERSITY,
PRINCETON, NEW JERSEY,
Jan. 21, 1957

[1] Vol. 46, p. 785–786, 1951.

El Vanadio en el Perú. By SILVIO HERNANDEZ AQUIJE. Minería (Organo del Instituto de Ingenieros de Minas del Peru), Lime. Año 2, No. 5, p. 19–30; No. 6, p. 31–49, 1954; Año 4, No. 8, p. 21–22; No. 9, p. 23–29, 1955.

Little new material descriptive of the world's richest vanadium deposit, Minasragra in Peru, has appeared since Hewett's excellent paper of 1909. From then until the recent period of inactivity, mining revealed much new information, particularly in regard to the behavior of the orebody in depth. Therefore, a recent series of papers in Spanish published in a journal that has less wide distribution than it deserves seems worth a rather detailed resumé.

The earlier papers of the series are concerned with the vanadium bearing asphaltite deposits that are so widely distributed through the Peruvian Andes. These deposits consist of fissures, or, in some cases, openings between beds, mostly of Lower Cretaceous limestone, filled with asphaltite whose content in V ranges from 0.3 to 1% or more in places. On burning, the V remains in the ash; in general the lower the percentage of ash the higher its V content which reaches 54% in the richest sample cited.

The Minasragra deposit although quite different from the asphaltites and much richer is linked to them by the common association of vanadium with carbonaceous material and by structural form. Contrary to some descriptions, it is fissure-like and transects the enclosing beds of red shale. The author distinguishes clearly between the primary deposit and the products of supergene alteration.

The primary deposit has the form of a wedge or an inverted cone, elongated in plan, zoned in such a manner that the outer portion adjoining the walls is quisqueite, a lustrous hydrocarbon, within which is natural coke enveloping central shoots of patronite, a vanadium sulfide of doubtful composition. At the surface the cone was lenticular in plan, measuring 350 × 28 feet. Downward it diminished in size until at the 200 level the "coke" had entirely disappeared; the patronite-bearing area was much reduced in size and diluted by material called "veta madre" (to be described later). The quisqueite continued and in one place extended as a tail to 40 feet below the 300 level or a depth of 253 feet below the surface.

This main body of ore and carbonaceous matter lies within an envelope of "bronce," consisting of shale impregnated with vanadium sulphide and some quisqueite. The bronce grades outward from dark gray shale through gray to brown shale into the surrounding red shale of the country. At the surface it occupies an area 850 × 180 feet, diminishing at depth to a vertex at about 300 feet.

The secondary mineralization results from weathering whereby vanadium sulphide has behaved much as would copper sulphide except that the minerals reprecipitated at depth were vanadates and vanadium oxides instead of sulphides. With depth, gypsum, derived from reaction of sulphuric acid with calcareous rocks, increases in amount and the supergene vanadium minerals are species increasingly soluble in sulphuric acid. Successive depth-zones are described as follows:

Upper 20 feet: Vanadates and oxides, red, blue, green and yellow but chiefly red and gray. Area 580 × 100 ft.

Next 46 feet: Oxides and vanadates, chiefly green.

Lower 167 feet: Oxides and vanadates dark green or gray. Gypsum is abundant. Maximum area 377 × 82 ft.

The material of this deepest zone is what is called "veta madre."

In its palmy days, the orebody yielded many thousand tons of a shipping product grading better than 6% V and by 1934 after nearly a quarter of a century of operation the reserves were estimated by W. Spencer Hutchinson as containing

70 million pounds of vanadium. All that is left now of this fabulous deposit is a hole 850 × 380 feet in area and 210 feet deep with the remnant of a vein which, according to the author, does not exceed 10,000 tons of 1.75% V content.

The July, 1955 installment of the paper contains half a dozen plans and sections of Minasragra drawn to scale.

In discussing origin, the author visualizes the vanadium-bearing carbonaceous material as asphaltite intruded into the fissure in a molten condition. Although a porphyry dike strikes roughly at right angles to the deposit, he believes that it antedates the ore as (1) it is strongly altered, particularly where it is adjoined by ore, and (2) it has been displaced by faulting which does not disturb the orebody. Therefore, he dismisses the possibility that it has metamorphosed the orebody or supplied any vanadium to it. He considers, however, that this or other intrusive material may have softened an original body of asphaltite and facilitated its emplacement in its present position.

As a matter of fact, although there are a number of intrusive bodies in the neighborhood it seems improbable that the vanadium was derived from them, as the numerous hydrothermal silver-base metal deposits associated with similar intrusives in the region are notably poor in vanadium minerals even in zones of oxidized lead ores. On the contrary, the association of vanadium with carbonaceous material points to a source in common with the asphaltites and suggests that Minasragra is an oversized and modified asphaltite deposit. Driving off the volatiles, presumably by the heat of intrusives, would account for coking of the material and thus accomplish some concentration of vanadium. The segregation of patronite in the center of the body remains to be explained.

HUGH MCKINSTRY

HARVARD UNIVERSITY,
Jan. 18, 1957

International Tin Study Group—The Statistical Year Book 1956. Pp. 284. The Hague, 1956. Price $5.60.

The Year Book is divided into two parts: Part I concerns Tin and Tinplate, Part II deals with the canning industry and trade. The book contains all available information for 110 countries, on the mine and smelter production, and trade of tin-in-concentrates, tin metal, tinplate, solder, and alloys. Most countries are also covered in the canning part. The book includes also six detailed sections on The Position of the International Tin Agreement, the World Tin Position 1946–55, the Assessment of Tin Consumption, the Recent Technical Developments in the Use of Tin (prepared by the Tin Research Institute), the World Tinplate Industry and the Canned Food Industry and Trade. As in the case of its predecessors, it is a very informative book.

R. JOIN

December, 1956

Sedimentary Rocks. 2nd Edit. By F. J. PETTIJOHN. Pp. 718; pl. 40; figs. 173; tables 119. Harper & Brothers, New York, 1957. Price, $12.00.

The first edition of "Sedimentary Rocks," published eight years ago, received excellent notices and has since become recognized as a basic text throughout the English-speaking world. It remains the only major postwar text concerned primarily with the description of the mega- and microscopic features of sedimentary rocks. The present volume is some 30 per cent larger than the first edition and the

number of plates and line drawings have been proportionately increased. The continued use of collotype plates for microphotographs, hand specimens, structural features and field exposures is fully justified by the excellence of the forty full-page plates used.

The subject matter falls naturally into three sections. The first four chapters deal with the general aspects of sedimentary rocks—texture, composition and structures. Grain size, shape, fabric and texture are all fully treated, with the accent on classification and statistical analysis. Chapter 3 describes the chemical and mineralogic composition of sediments, their detrital minerals and the chemical precipitates. This is followed by a discussion of structures of primary, secondary and organic origin, ranging in scale from bedding down to micro-organic structures.

The six chapters on the description of sedimentary rock types form the second and largest part of the book. After an introductory chapter on classification, the clastic rocks are described in turn, followed by the carbonate and non-clastic sediments. The dual bases of grain size and chemical composition are combined to give a framework convenient for descriptive purposes and following current usage. Throughout these chapters description is backed by genetic considerations and controversial subjects are summarized fairly. The excellent microphotographs that illustrate this section are some of the clearest the writer has seen.

The final chapters of the book deal with the more general and theoretical aspects of sediments. The two chapters on Provenance and Dispersal relate the features of sedimentary rocks to source, process and cycle. The significance of mineral stability, grain size and shape is especially discussed and there is a new section on the mapping and interpretation of vector properties of sediments. Chapter 13 is another new section describing the relation between environment of deposition and the characters of the sediment and then relates sedimentary facies to tectonic environment. After a chapter on the processes of lithification and diagenesis, the book ends with "The Historical Geology of Sediments," containing a discussion of the geochemical evolution of sediments and leading the reader quite logically into the subject of Historical Geology itself.

Throughout its length the large scope of this book is apparent—description and genesis go hand in hand and each subject is exhaustively discussed with a minimum of dogma and a maximum of clarity. The abundance of tabulated data and the extensive bibliographies that follow each important section make this book not only an excellent text but a valuable reference. With this growth in size, Pettijohn's "Sedimentary Rocks" moves from a book to a tome. Certainly there is no similar text in the English language that can match it for size, scope, and volume of contents.

LAURENCE S. PHILLIPS

YALE UNIVERSITY,
NEW HAVEN, CONN.,
March 19, 1957

BOOKS RECEIVED

V. RAMA MURTHY

The Limestones of Scotland. Chemical Analyses and Petrography. A. MUIR, H. G. M. HARDIE, R. L. MITCHELL, and J. PHEMISTER. Pp. 150, pls. 4. Price, £1 1s. Department of Scientific and Industrial Research, Geological Survey of Great Britain, Edinburgh, 1956. *Compilation of chemical and spectrographic analyses of Scottish limestones with brief petrographic descriptions.*

The First One Hundred and Fifty Years. A History of John Wiley and Sons, Inc. Pp. 242. Price, $7.50. *A very well compiled history of John Wiley & Sons, Inc., outlining their publications in diverse fields in the past one hundred and fifty years. Each chapter is contributed by a noted man in the field.*

Radiation Safety and Major Activities in the Atomic Energy Programs, July–December 1956. Pp. 396. Price, $1.25. U. S. Government Printing Office, Washington, 1957.

Notes on Atomic Energy for Medical Officers. OFFICERS OF THE ROYAL NAVAL MEDICAL SCHOOL, ALVERSTOKE, HAMPSHIRE, ENGLAND. Pp. 169, pls. 12. Price, $4.75. Philosophical Library, Inc., New York, 1956. *Written for the purpose of providing basic knowledge of short and long-term effects of atomic explosion and dangers from other kinds of radiological warfare. A good introduction to the subject.*

Minerals Yearbook Area Reports, Volume III, 1953. FIELD STAFF OF BUREAU OF MINES. Pp. 1169. Price, $4.00. U. S. Government Printing Office, Washington, 1956. *Mineral data for each of the 48 states and chapters on the Territory of Alaska, the Territory of Hawaii and island possessions in the Pacific Ocean, and the Commonwealth of Puerto Rico.*

Geochemical and Petrographic Aspects of Mercury Ore Deposits. FRANK W. DICKSON and GEORGE TUNELL. Pp. 79, pls. 4, figs. 23. Dept. of Geology, Univ. of California, Los Angeles, 1955. *An exhaustive study of solubility variation of HgS in alkaline solutions at several temperatures, and a study of the relationship of mercury ore minerals to the gangue minerals associated with them, in order to determine the nature of ore-solutions and conditions of deposition.*

Report of the Committee on the Measurement of Geologic Time 1953–1954. JOHN PUTNAM MARBLE, CHAIRMAN. Pp. 193. Price, $1.75. National Academy of Sciences—National Research Council, Washington, 1955.

Structural Geology Laboratory Manual. JOHN C. LUDLUM and JOHN M. DENNISON. Pp. 115, figs. 47. John C. Ludlum, Morgantown, West Virginia, 1957. *Class-room manual, written clearly, and includes exercises at the end of each chapter. Commendable emphasis seems given to mathematical analysis of structural data.*

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Bull. 30. The Geology of the Superficial Deposits and Coastal Sediments of British Guiana. D. BLEACKLEY. Pp. 46, figs. 14. Price, $3.00. British Guiana Geological Survey, Georgetown, Demerara, 1956.

Foraminifera from the Wagonwheel Formation Devils Den District, California. HUGH P. SMITH. Pp. 108, pls. 16. Price, $1.50. University of California Press, Berkeley and Los Angeles, 1956.

Sixth Annual Report, 1955, National Advisory Committee on Research in the Geological Sciences. Pp. 57. Price, 50 cts. Geological Survey of Canada, Ottawa, 1956. *Review of activities and progress made in the geological sciences in the year 1955–56.*

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The Economic Geology of the Stirling and Clackmannan Coalfield Scotland Area North of the River Forth. E. H. FRANCIS. Pp. 36, figs. 13, tbls. 2. Price, 5 sh. Geological Survey of Great Britain, London, 1956.

Rept. of Investigations 199. Conodonts from the Chester Series in the Type Area of Southwestern Illinois. CARL B. REXROAD. Pp. 43, pls. 4, figs. 21, tbl. 1. Illinois Geological Survey, Urbana, 1957.

Contribution 47. The Structural History of the Northern Part of Iwate Prefecture. NOBU KITAMURA, TAMIO KOTAKA, and SHÔZÔ HAYASAKA. Pp. 20, figs. 10. Institute of Geology and Paleontology Tohuku University, Sendai, Japan, 1956.

Official Oil in North Dakota. Production Statistics, Second Half 1956. Pp. 31. North Dakota Geological Survey, Grand Forks, 1957.

Bull. 55. Geology of Tuscarawas County. RAYMOND E. LAMBORN. Pp. 269. Maps. Price, $3.00. Division of Geological Survey, Ohio State University, Columbus, 1956. *A description of the physical Geology of Tuscarawas County; surface features and their history, bed rock, including limestone, sandstone, shale, and coal, and their economic importance; and oil and gas production and possibilities.*

Special Projects Series, Pub. 7. Geology and Coal Resources of the Argao-Dalaguete Region, Cebu. HARLEY BARNES, CRESENCIO P. JONGCO, GENEROSO C. LAZAGA, and others. In two parts. Pp. 52, pls. 8, figs. 2, tbls. 5. Price, $3.00. Philippine Bureau of Mines, Manila, 1956.

Annual Report of the Tanganyika Geological Survey Department 1955. Pp. 28. Price, 3 sh. Dar es Salaam, 1956.

Publication 5704. Pelecypoda from the Type Locality of the Stone City Beds (Middle Eocene) of Texas. H. B. STENZEL, E. K. KRAUSE, and J. T. TWINING. Pp. 237, figs. 31, pls. 22. Price, $3.75. University of Texas Bureau of Economic Geology, Austin, 1957.

Union of South Africa—Geological Map. Scale 1:1,000,000, 1955 (1957). *4 sheets 28 × 38 inches, colored.*

Memoir 66. Cenozoic Foraminifera of Micronesia. SHOSHIRO HANZAWA. Pp. 163, pls. 41, figs. 12, tbls. 7. Geological Society of America, New York, 1957. *Detailed study of the larger Foraminifera found in the Priabonian, Aquitanian, Burdigalian, Pliocene?, and Pleistocene sediments of Micronesia.*

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Facultad de Ingenieria Y ciencias Exactas Fisicas y Naturales, Universidad Nacional de Cuyo, San Juan, Republica Argentina, 1954.

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Acta Cuyana de Ingenieria. Vol. 1, No. 7. Formulas para las Areas y Volumenes Limitados por los Contornos y Superficies Cerradas. SERGIO SISPANOV. Pp. 12.

Bureau of Mineral Resources, Geology and Geophysics—Australia, 1955–1956.

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Geological Rept. 68. McGill Area, Papineau, LaBelle and Gatineau Counties. E. AUBERT DE LA RÜE. Pp. 21, pls. 4. Map in pocket. The Mining Industry of the Province of Quebec in 1955. Pp. 131, pls. 4, tbls. 54.

Idaho Bureau of Mines and Geology, Moscow, 1956–1957.

Pamphlet 111. Economic Evaluation of Phosphate and Other Minerals in Southern Idaho. JAMES F. MCDIVITT. Pp. 48, tbls. 6.

Pamphlet 112. Geology and Mineral Resources of the Baker Quadrangle, Lemhi County, Idaho. ALFRED L. ANDERSON. Pp. 71, pls. 7, fig. 1.

Indiana Department of Conservation, Bloomington, 1956.

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Geological Survey of Japan—Hisamoto-cho, Kawasaki-shi, 1956.

Explanatory Text of the Geological Map of Japan. Saruru (Kushiro-71). SACHIO IGI and MITSUO HATA. Pp. 34, figs. 2, tbl. 1. *Geologic map, 1:50,000.*

Explanatory Text of the Geological Map of Japan. Ominagahama (Kyōto-6). HIROSHI ISOMI. Pp. 57. *Geologic map, 1:50,000.*

Explanatory Text of the Geological Map of Japan. Karatsu (Fukuoka-59). ISAMU KOBAYASHI, ISAO IMAI, and KAZUNORI MATSUI. Pp. 69, figs. 16, tbls. 12. *Geologic map, 1:50,000.*

Rept. 169. Natural Gas Accumulation in Japan. KATSU KANEKO. Pp. 62, figs. 31, tbls. 16.

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U. S. Geological Survey—Washington, D. C., 1956–1957.

Bull. 1019-D. Bibliography of Iron Ore Resources of the World (To January 1955). GWENDOLYN W. LUTTRELL. Pp. 187–371. Price, 65 cts.

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Bull. 1021-K. Geology of the Atomic Energy Commission Nevada Proving Grounds Area, Nevada. MIKE S. JOHNSON and DONALD E. HIBBARD. Pp. 333–384; pls. 32–33; figs. 57. Price, $1.00. *Stratigraphic and structural study of the area.*

Bull. 1030-K. Uranium Deposits in Oolitic Limestone Near Mayoworth, Johnson County, Wyoming. R. R. GUILINGER and P. K. THEOBALD. Pp. 335–342, figs. 62–66. Price, 15 cts. *Metatyuyamunite occurs as coatings on joint surfaces and as replacement of oolites; the grade of ore is not of economic importance at present.*

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Prof. Paper 274-J. Fossils from the Eutaw Formation Chattahoochee River Region, Alabama-Georgia. LLOYD WILLIAM STEPHENSON. Pp. 227–250, pls. 38–45, fig. 30. Price, 70 cts.

Prof. Paper 280-A. Geology of Saipan Mariana Islands, Part 1. General Geology. PRESTON E. CLOUD, JR., ROBERT G. SCHMIDT, and HAROLD W. BURKE. Pp. 126, pls. 25, figs. 10, tbls. 11. Price, $3.00.

Water-Supply Paper 1360-E. Geology and Groundwater Resources of the Kaycee Irrigation Project Johnson County Wyoming. F. A. KOHOUT. Pp. 321–374, pls. 22–23, figs. 46–54, tbls. 1–7. Chart in pocket.

Water-Supply Paper 1412. Water Consumption by Water-Loving Plants in the Malad Valley Oneida County, Idaho. R. W. MOWER and R. L. NACE. Pp. 33, pls. 2, figs. 8, tbls. 11. Price, 55 cts.

SCIENTIFIC NOTES AND NEWS

R. C. VORHIS, U. S. Geological Survey, has been assigned to Tripoli, Libya, following his evacuation in October from Cairo, Egypt. He will be engaged in ground-water studies sponsored by the International Cooperation Administration.

JOSEPH L. GILLSON gave the 1957 Bownocker Lectures in the Department of Geology, Ohio State University, Columbus, on March 5 and 6. He presented two afternoon talks on "A Geologist Works with Industrial Minerals," and an illustrated evening lecture entitled "Foreign Exploration for the Wonder Metal, Titanium."

FREDRICK C. KRUGER, formerly with Reynolds Metal Co., has been appointed chief geologist for International Minerals and Chemical Corporation, Chicago, and will be primarily responsible for geological appraisals and the evaluation of new mineral discoveries.

GEORGE W. RUST, formerly senior geologist with Bear Creek Mining Co., has joined the staff of the Pacific Coast Company as Director of Mining and Exploration, with headquarters at Room 1900, Russ Building, San Francisco, California. This old company with highly diversified interests, is broadening its scope of activities in the mining field.

The names of the STANOLIND OIL AND GAS CO. and the STANOLIND FOUNDATION have been changed to the PAN AMERICAN PETROLEUM CORPORATION and the PAN AMERICAN PETROLEUM FOUNDATION, INC.

When authors submit papers describing investigations in which X-ray powder measurements were made, but omitting the actual X-ray data, it is requested that these data be submitted to G. W. Brindley, Editor of the Joint Committee on Chemical Analysis by Powder Diffraction Methods for possible inclusion in the "X-ray Powder Data Field," published by the A.S.T.M. The data should contain accurate listings of "d" values and "intensities of reflections," hkl indices and lattice parameters if known, radiation used, type of X-ray recording employed, method of estimating intensities (visual, photometric, geiger-counter), plus other relevant information. The Pennsylvania State University, University Park, Pennsylvania, U.S.A.

The NEW MEXICO GEOLOGICAL SOCIETY will hold its Eighth Annual Field Conference in the Durango-Silverton-Ouray area of southwestern Colorado, September 5, 6, and 7, 1957.

The NATIONAL SCIENCE FOUNDATION announced today that the Corps of Engineers, U. S. Army, has undertaken acquisition of the site for the Foundation-supported radio astronomy observatory at Green Bank, West Virginia. The Engineers, acting on behalf of the Foundation, have established a field office for this purpose at Marlinton, West Virginia. The new research center for radio astronomy will be constructed and operated for the use of the Nation's scientists by Associated Universities, Inc., under contract to the Foundation.

McGILL UNIVERSITY, Department of Geological Sciences, is offering a new two-year sequence of courses leading to a degree of Master of Science in Mineral

Exploration. The new degree is designed to provide advanced professional training in geology, mining, metallurgy, physics, and chemistry for students entering the mineral exploration field. The new technical methods of exploration that have recently been developed require more basic training than can be given in undergraduate work. The course may be entered with the Bachelor's degree in Mining Engineering or the B.Sc. degree with Honors in Geology. Students who complete the M.Sc. (Applied) sequence with high standing may continue towards the normal Ph.D. (Geology) degree.

E. A. SWEDENBORG, retired from the U. S. Geological Survey in December after 30 years, has been appointed Wyoming State Minerals Supervisor, succeeding GEORG JARVIS who resigned February 1.

FRANK P. KNIGHT has been named director of the Arizona Department of Mineral Resources.

VINCENT D. PERRY, chief geologist for Anaconda Company since 1948, has been elected a vice-president of the company.

C. A. LEHNERTZ, staff geologist with Doeringsfled, Amuedo & Ivey, of Denver, is on a six-month assignment to Jamaica where he is working on a photogeological survey of the island.

DONALD W. LINDGREN has been named chief mining geologist for the Northern Pacific Railway Co., with headquarters at St. Paul, Minn., where he will supervise a staff of mining geologists engaged in the exploration and development of the mineral resources of the company's large land holdings, exclusive of oil and gas.

EUGENE CALLAGHAN, director of the New Mexico Bureau of Mines and Mineral Resources for the past 7½ years, has been appointed chief geologist in charge of exploration for Haile Mines, Inc., with field headquarters in Salt Lake City.

ALVIN J. THOMPSON will succeed Eugene Callaghan who resigned as director of the New Mexico Bureau of Mines and Mineral Resources. Mr. Thompson was formerly head of the mining department of the bureau.

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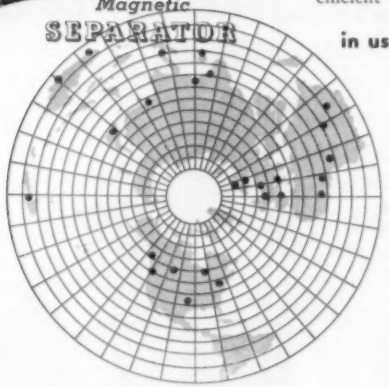

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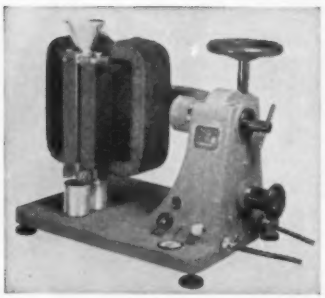

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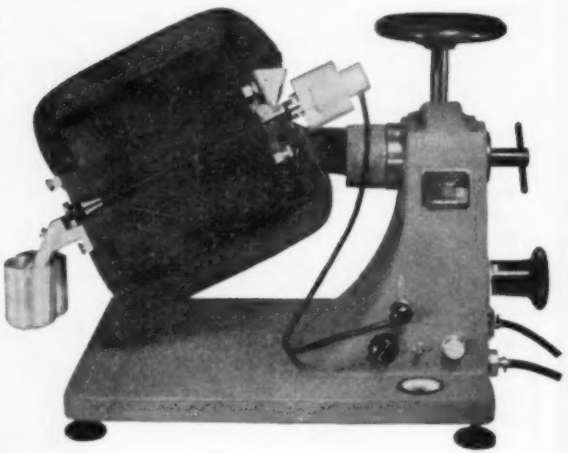

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